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CARCINOGENS AND MUTAGENS IN AMBIENT AIR PARTICULATE MATTER: SOURCES AND TRENDS IN CONTRA COSTA COUNTY

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ABSTRACT

Many mutagens and carcinogens are known to be present in urban community air. Extensive chemical and biological characterization of these atmospheric pollutants is essential if accurate risk assessments are to be made and effective control strategies developed. This report describes progress in three areas of this complex environmental problem: 1. the development of more sensitive methods for measuring aerosol mutagens, 2. the identification of sources of mutagens and 3. the analysis of trends in mutagen and polycyclic aromatic hydrocarbon (PAH) levels in particulate organic matter (POM).

- A highly sensitive version of the Ames Salmonella test, called the microsuspension test, was applied to measure the mutagenic activity in organic extracts of community aerosols. Application of the microsuspension Ames test made possible high resolution diurnal studies of mutagenicity in small air samples of only 2 hours duration. Diurnal variations in mutagenic density (revertants/m^3) of more than a factor of 10 were observed and these variations were highly correlated with fine fraction lead (Pb) in a pilot field study. The test can be applied in future studies were sample mass is a limiting factor.

- The origins of mutagens in POM were investigated further by sampling in Contra Costa County during six seasonal pollution episodes, each of 36 hours duration, in 1982-1984. Samples were collected at four locations (Richmond, Martinez, Concord, Pittsburg) and analyzed for mutagenic activity in the Ames test, for PAH, oxyanions (NO_3^-, $SO_4^=$), pollutant gases (CO, NO, NO_2, O_3, SO_2) and elemental source tracers (including Pb, Br, Ni, Fe and K). Diurnal, geographic and seasonal comparisons were made. Statistical techniques, including principal component (factor) analysis, were used to explore relationships between aerosol mutagens, PAH and source tracers. The results confirmed earlier observations and provided some new insights into the sources of aerosol mutagens.

    (i) Several lines of evidence indicate that some mutagenic aerosols are primary automotive pollutants emitted directly into the atmosphere.

a. In this present and previous Contra Costa studies, mutagenic density and PAH were significantly positively correlated with fine fraction ($< 2.5\ \mu md_a$) Pb and/or Br, both derived primarily from motor vehicles.

b. Chemical analysis by other investigators has identified mutagens (various PAH and nitroarenes) in on-road vehicle particulate emissions, as well as other combustion source particulate matter.

c. Studies of upwind-downwind freeway data in Los Angeles by Statewide Air Pollution Research Center (SAPRC) scientists have demonstrated an incremental burden of direct mutagens in aerosol attributable to freeway traffic. The amount was comparable to the area wide background mutagen density.

(ii) Many results suggest that some mutagens behave as secondary aerosols. The hypothesis that some mutagenic aerosols are formed in the atmosphere is supported by the following evidence:

a. During pollutation episodes in Contra Costa County, mutagens were positively correlated with NO_3^-, assumed to be a secondary aerosol tracer. The association of mutagenicity with NO_3^- occurred areawide.

b. SAPRC scientists observed that ratios of mutagen densities (rev/m^3) to CO were generally higher at Riverside, California, a downwind receptor site, than at El Monte, an intermediate receptor site in the Los Angeles basin. Since CO is an unreactive combustion emission, the mutagen density/CO ratio takes into account variations in emissions and atmospheric dispersion. Higher ratios at Riverside suggest atmospheric mutagen formation during aerosol transport from Los Angeles.

c. The ratios of mutagenic densities to Pb which we have measured in Contra Costa County in this and a previous study were highest

during summer episodes when the prevailing atmospheric conditions (i.e. hot, dry, stagnant) favored chemical transformations. Since Pb, like CO is an unreactive emission, the mutagenic density/Pb ratio should take into account variations in automotive emission profiles and dispersion. Thus the high ratios during episodes in August 1981 and September 1983 may reflect atmospheric mutagen formation.

d. Smog chamber studies have demonstrated the formation of nitro-PAH mutagens. Mutagenicity of some nitro-PAH's exceed the mutagenicity of the parent PAH by several orders of magnitude in laboratory analysis. Some of these highly mutagenic nitro-PAH's are known to be primary pollutants emitted by various combustion sources. However chamber studies have also shown that irradiation of mixtures of atmospheric hydrocarbons, nitric acid (HNO_3) and reactive gases (NO_2, O_3) can lead to mutagen formation. Thus some hydrocarbons may be converted to secondary mutagenic products under simulated atmospheric conditions.

e. Measurements in a nitroreductase mutant indicate the likely presence of nitroarene mutagens. Less than 10% of the total mutagenicity in ambient air samples is due to identified PAH. Thus most of the mutagenicity remains to be explained in chemical terms. A substantial proportion of this excess mutagenicity may be due to highly mutagenic nitroarenes and derivatives, which are not only ubiquitious primary pollutants but may also be derived from secondary atmospheric transformations. We infer that such compounds were probably major contributors to the mutagenicity of Contra Costa aerosols from the fact that mutagenic activities of aerosol extracts were two to three times lower in a Salmonella strain (TA98NR) deficient in an enzyme required for some mono-nitroarene activation, than in the standard tester strain (TA98).

f. Finally, measurement artifacts confound the secondary mutagen hypothesis. The positive correlations of mutagenic density with NO_3^-, and the demonstration that mutagenic organic compounds can be formed under simulated atmospheric conditions support the hypothesis of secondary formation of mutagenic aerosols in the atmosphere. The association between mutagens and NO_3^- can be influenced by HNO_3 artifacts produced by sampling on glass fiber filters. There are two concerns. Gas phase HNO_3 can bind to glass fiber and artificially increase apparent particulate NO_3^- concentrations. More importantly, gas phase HNO_3 may catalyze chemical tranformations of PAH to produce highly mutagenic nitroaromatic compounds during sample collection on glass fiber. The significance of these potential artifacts cannot be assessed accurately at present.

(iii) For the first time in Contra Costa County, industrial contributions to mutagenic aerosols were suggested by significant positive correlations between mutagenic density and S (both fine fraction S and SO_2) at Richmond and Martinez. Sulfur oxides are major air pollutants in the vicinity of large oil refineries and chemical plants in Contra Costa County. The major industrial sources of SO_2 are refineries in Richmond (Chevron), Martinez (Shell, Tosco) and Benicia (Exxon) and a chemical plant in Rodeo (Union).

- Routine collection and analysis of 4 month seasonal composite filter samples was carried out in Contra Costa County between 1979-1984. The three periods were Nov.-Feb., March-June and July-Oct. These periods approximate the three meteorological seasons in the area.

This monitoring effort demonstrated that levels of most aerosol pollutants including mutagens and PAH, were highest in the winter (Nov.-Feb.).

A prime goal of the monitoring was to detect any time trends which may have occurred. Monitoring did indeed reveal a positive trend in the concentration of mutagenic aerosols, despite decreasing or constant levels of the other pollutants

measured. The annual average increased from 5 revertants/m^3 in 1979-80 to 19 revertants/m^3 in 1983-84. A three to four-fold increase in mutagenic density (from 8 revertants/m^3 to 27 revertants/m^3) was observed over the five winter seasons. Values in the spring increased from 2 to 18 revertants/m^3 while summertime values increased by more than a factor of two from 5 to 13 revertants/m^3. Further monitoring is needed to determine the persistence of these trends.

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We wish to express our appreciation to the following staff of the Air and Industrial Hygiene Laboratory who provided consultation and performed chemical determinations: S. Twiss, W. Wehrmeister, A. Cartano, Z. Ilejay, F. Boo, N. Fansah, E. Jeung, E. Hoffer, and A. Alcocer. We also thank R. Giaque of the Lawrence Berkeley Laboratory LBL who performed the trace element analysis and J. Jaklevic and B. Loo, (LBL), who provided the Automatic Dichomotous Samplers.

Finally, we thank Project Officer C. Unger for his direction and encouragement.

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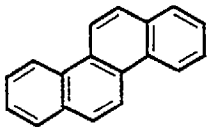
CHAPTER I
PROJECT SUMMARY

A. Introduction and Statement of the Problem

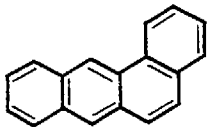
A variety of chemical mutagens and carcinogens are known to be present in particulate matter in urban community air (1-9). Recent epidemiological conclusions place the number of cancer deaths in the U.S. due to environmental pollution (air, water and soil) at 8,000 annually (7). It has been estimated that outdoor community air pollution alone may be the cause of about 15% of the present lung cancer cases among non-smoking residents in Contra Costa County (8), neglecting contributions from indoor air pollution. Thus it may be argued that exposure to mutagens and carcinogens in outdoor community air remains a significant public health concern which warrants continued monitoring and investigation.

Carcinogenic chemicals found in air particulate matter include certain polycyclic aromatic hydrocarbons (PAH), such as benzo(a)pyrene (BaP) and benzo(k)fluoranthene (9). Structures of these and other PAH are shown in Figure I-1. However, these chemicals constitute only a small fraction of the total carcinogenic potential. Organic extracts of ambient particulate organic matter (POM) are significantly more carcinogenic and mutagenic than expected on the basis of the amounts of the measured chemicals present (10,11). The excess mutagenicity, as defined by the Ames test (12), reflects a significant gap in our knowledge. The gap may be due to our inability until recently to measure small concentrations of carcinogenic and highly mutagenic nitroarenes (e.g. 1-nitropyrene) (13) which are present in diesel exhaust particulates (14) and urban air (15,16,17). These compounds probably account for a significant portion of the observed mutagenicity of urban air particulate matter.

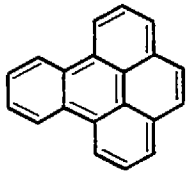
However, the measurement of mutagenicity in a given geographical area and even the chemical identification of the mutagens present are of limited public health value unless the major sources of mutagenicity can be identified and controlled. With few exceptions, the sources of mutagens and carcinogens in air particulates are unknown. In Contra Costa County, for example, PAH



Chrysene



Benz(a)anthracene



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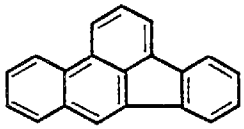

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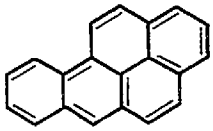
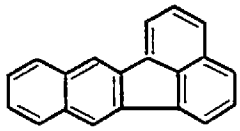
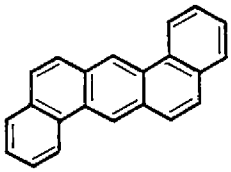


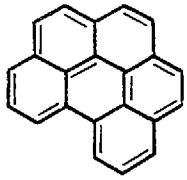
Benzo(a)pyrene



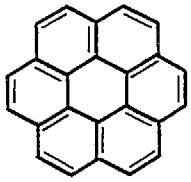
Benzo(k)fluoranthene



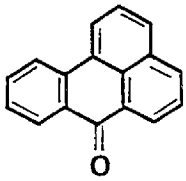
Dibenz(a,h)anthracene



Benzo(ghi)perylene



Coronene



7-H-benz(d,e)anthracen-7-one

FIGURE I-1 STRUCTURE AND NOMENCLATURE OF 10 POM's

appeared to be derived principally from vehicular emissions but these compounds account for less than ten percent of measured mutagenicity (6). Until recently the principal sources of mutagens remained obscure.

Concern about sources of mutagen led to a previous study of mutagens in Contra Costa County carried out by the Air and Industrial Hygiene Laboratory (AIHL) under contract to the California Air Resources Board (CARB) (18). The AIHL-CARB project provided the background to the present investigation. The previous study measured the mutagenic activity of ambient aerosol extracts and quantitated a number of organic compounds which contribute to the mutagenicity. Air sampling was carried out during three seasonal pollution episodes in 1981-1982. Solvent extracts of POM were analyzed for mutagenic activity in the Ames Salmonella test and for polycyclic aromatic hydrocarbons (PAH) by high pressure liquid chromatography (HPLC) with fluorescence detection. Pb, Ni, Fe, NO_3^-, $SO_4^=$, O_3, CO, NO, NO_2, and SO_2 were also measured. Diurnal variations in mutagenicity and chemical pollutant concentrations were compared. Mutagenicity was found to be consistently and strongly associated with lead-containing fine particles ($< 2.5\ \mu md_a$) emitted primarily by gasoline powered vehicles without catalysts. During a winter inversion, possible contributions to PAH from residential wood combustion were also noted.

The previous study also revealed positive correlations between mutagenicity and nitrates during a summer pollution episode. We speculated earlier that this correlation could reflect chemical formation of nitro PAH compounds in the atmosphere and suggested that in some respects, the formation of mutagenic aerosols was similar to the formation of photochemical oxidant; the effects of atmospheric dispersion and transformation both need to be considered. Since this suggestion could have policy implications for control of oxides of nitrogen, further investigation was required.

B. Project Objectives

The present study was designed to followup these earlier leads by addressing three topics which are critical to a further understanding of mutagens and carcinogens in community air: 1) the validation of a more sensitive method

for measuring aerosol mutagens, 2) the identification of the sources and the possible routes of atmospheric chemical formation of aerosol mutagens and 3) the investigation of seasonal variations and the recent trends in airborne mutagen and PAH concentrations in Contra Costa County between 1979 and 1984.

C. Experimental Approach

The research carried out under this and the previous CARB-contract, (18), employed a similar experimental approach and methods. The abbreviations of air pollutants and methods used for their collection and analysis are listed in Tables I-1 and 2.

1. Application of the Salmonella Microsuspension Procedure to the Measurement of Mutagenicity in Air Particulate Matter

A simple and highly sensitive version of the Salmonella liquid incubation assay (19) was validated using pure chemical mutagens and then applied to the measurement of air particulate mutagenicity. These applications included diurnal studies carried out at several locations. In one experiment, selected criteria pollutants (Pb, NO_2, O_3, SO_2) were sampled concurrently, to provide information about sources of mutagens.

2. Intensive Sampling for Mutagen and PAH Source Identification

A second focus of the present effort was on source identification. Six pollution periods were sampled intensively to investigate sources of mutagens and PAH. The sampling and analytical plan for source identification is shown in Table I-3. Separate "day" (0600-1800) and "night" (1800-0600) sampling was carried out over 36 hour episodes. Episodes were stable meteorological periods, when concentrations of pollutants were relatively high. Samples were collected at four locations (Richmond, Martinez, Concord and Pittsburg) in the northern industrialized portion of Contra Costa County (Figure I-2). The northern section of the county contains heavy industry including five major petroleum refineries and many chemical

TABLE I-1

ACRONYMS FOR AIR POLLUTANT VARIABLES USED IN THE ANALYSIS AND INTERPRETATION OF CONTRA COSTA DATA

TSP	Total Suspended Particulate Mass	$\mu g/m^3$
$SO_4^=$	Sulfate Mass	$\mu g/m^3$
NO_3^-	Nitrate Mass	$\mu g/m^3$
ORG	Benzene Soluble Organics	$\mu g/m^3$
BAP	Benzo(a)pyrene	ng/m^3
BKF	Benzo(k)fluoranthene	ng/m^3
BGP	Benzo(ghi)perylene	ng/m^3
COR	Coronene	ng/m^3
BZO	Benzanthrone (all isomers)	ng/m^3
PBF	Fine fraction Lead	ng/m^3
BRF	Fine fraction Bromine	ng/m^3
FEF	Fine fraction Iron	ng/m^3
SIF	Fine fraction Silica	ng/m^3
KF	Fine fraction Potassium	ng/m^3
ZNF	Fine fraction Zinc	ng/m^3
SF	Fine fraction Sulfur	ng/m^3
CLF	Fine fraction Chlorine	ng/m^3
M398PS9	Revertants per m^3 in TA98+S9	rev/m^3
M398MS9	Revertants per m^3 in TA98-S9	rev/m^3
M398NRM	Revertants per m^3 in TA98NR-S9	rev/m^3
NR/98M3	Revertants per m^3 in TA98NR/TA98	-
ORG98PS9	Revertants per ORG in TA98 + S9	$rev/\mu g$
ORG98MS9	Revertants per ORG in TA89-S9	$rev/\mu g$
O_3	Ozone	pphm
CO	Carbon Monoxide	ppm
NO	Nitrogen oxide	pphm
NO_2	Nitrogen dioxide	pphm
SO_2	Sulfur dioxide	pphm

TABLE I-2

METHODS USED FOR COLLECTION AND ANALYSIS OF PARTICULATE AND GASEOUS AIR POLLUTANTS

Pollutant	Collection (Medium)	Analysis
TSP	Hi-vol (glass fiber)	Gravimetric
$SO_4^=$	Hi-vol (glass fiber)	Turbidimetric
NO_3^-	Hi-vol (glass fiber)	Colorimetric
ORG	Hi-vol (glass fiber)	Benzene extraction
PAH	Hi-vol (glass fiber)	HPLC - fluorescence
Mutagens	Hi-vol (glass fiber)	Standard Ames test
	Dichotomous (Teflon)	Microsuspension Ames test
Trace elements	Dichotomous (Teflon)	X-ray fluorescence
	Hi-Vol (glass fiber)*	" "
O_3	Dasibi, Model 1003-AH	Ultraviolet absorption
NO, NO_2	Thermal-electron, Model 14D	Chemiluminescence
CO	Bendix, Model 8301-5CA	Infrared absorption
SO_2	Thermal-electron, Model 43	Fluorescence

*Pb only.

TABLE I-3

SAMPLING AND ANALYTICAL PLAN FOR MUTAGEN SOURCE IDENTIFICATION

Day-Night Collection: 6 a.m. - 6 p.m.
6 p.m. - 6 a.m.

Collect Particulates on:

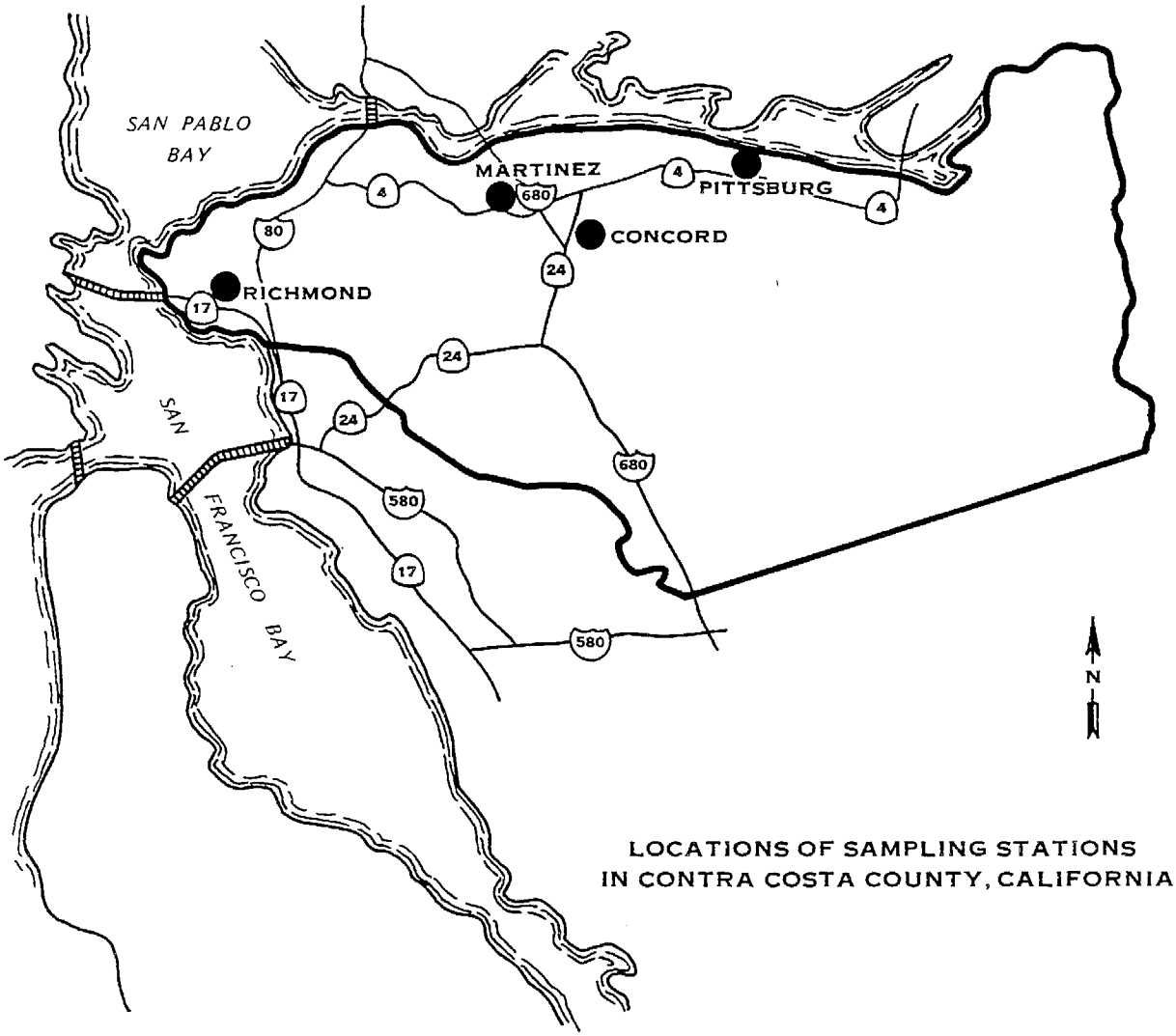
	Analytical Method	Agency Performing Analysis
Hi-vol #1 (glass fiber)		
Mass	Gravimetric	AIHL
Sulfate	Turbidimetric	AIHL
Nitrate	Colorimetric	AIHL
Lead	X-ray Fluorescence	AIHL
Organics	Benzene Extraction	AIHL
Hi-vol #2 (glass fiber) Refrigerate immediately		
Mutagenicity	Ames	AIHL
PAH	HPLC	AIHL
Dichotomous Samplers (membrane or Teflon filter)		
Multielemental analysis	Energy Dispersive X-ray Fluorescence	DHS-LBL

Collect Gas Data

NO_x	Chemiluminescence Photometry	BAAQMD
CO	Non-Dispersive Infrared Absorption	BAAQMD
SO_2	Fluorescence Photometry	BAAQMD
O_3	Ultraviolet Absoprtion	BAAQMD

Collect Meteorological Data

Wind direction Wind speed	BAAQMD



LOCATIONS OF SAMPLING STATIONS
IN CONTRA COSTA COUNTY, CALIFORNIA

FIGURE I-2

plants. Three of the stations (Richmond, Concord, Pittsburg) are part of the Bay Area Air Quality Management District (BAAQMD) network. Martinez was a temporary site, adjacent to a petrochemical refinery.

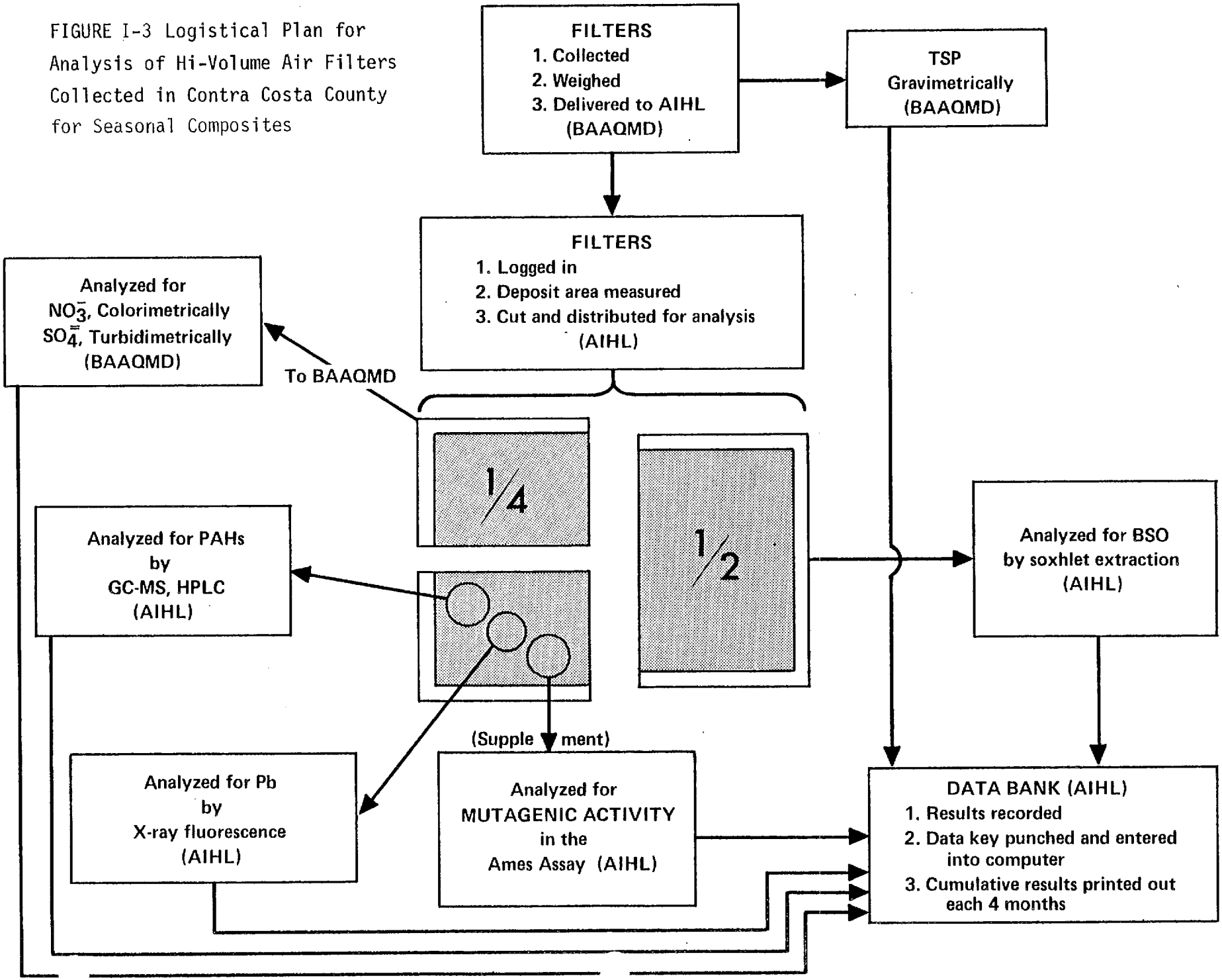
Each location had samplers to collect air particulate matter for analysis of mutagenicity, PAH, trace metals (including Pb, Ni, K, Si), NO_3^-, $SO_4^=$ and total mass. Gaseous pollutants (CO, SO_2, NO, NO_2, O_3) were also measured. At Martinez, wind speed and direction were obtained. Chemical and mutagenicity data were combined using simple and complex statistical methods in an attempt to identify sources of mutagens and selected PAH.

3. Collection and Analysis of Seasonal Composites

To determine seasonal variations and trends, samples were collected at the same three permanent stations of the BAAQMD network (Concord, Pittsburg and Richmond) used for intensive sampling. Hi-vol filter samples were collected every sixth day at each station for routine monitoring purposes and were analyzed for total suspended particulate (TSP), $SO_4^=$, NO_3^-, organics and Pb. A portion of each filter was composited for PAH and mutagenicity testing. Each station was composited separately. The logistical plan for analysis of hi-vol filters collected for seasonal composites is shown in Figure I-3. Filters from each of the three stations were composited over four-month intervals (July-October, November-February, March-June), to give composite samples for analysis. These periods approximate the three meteorological seasons in the San Francisco Bay air basin and also correspond with those used in our previous studies in Contra Costa County (18).

Samples collected during the period July 1982-October 1984 were composited and analyzed for PAH and mutagenic activity. When combined with results of previous studies, these provide a continuous data base of the concentrations of specific PAH and mutagenic activity in Contra Costa air particulate material collected over five years, since November 1979. Results of PAH and mutagenicity measurements in composite samples were also compared with TSP, NO_3^-, $SO_4^=$, Pb and total organics on a season-by-season basis.

FIGURE I-3 Logistical Plan for Analysis of Hi-Volume Air Filters Collected in Contra Costa County for Seasonal Composites



D. Summary of Findings

Efforts to validate and apply a highly sensitive version of the Ames test to air samples (Chapter II) yielded the following findings:

1. The 10 fold increased sensitivity of the "microsuspension" Ames test made possible high resolution diurnal studies of mutagenicity in small samples of only 2 hours duration.

2. Diurnal variations in mutagenic density (rev/m^3) of more than a factor of 10 were observed.

3. Diurnal variations in mutagenic density were highly correlated with fine fraction Pb, in a pilot field study.

4. The test can be applied in future studies where sample mass is a limiting factor.

Intensive episode sampling and analysis for source identification (Chapter III) confirmed earlier observations and provided now new insights into sources of aerosol mutagens.

1. Several lines of evidence indicate that some mutagenic aerosols are primary automotive pollutants emitted directly into the atmosphere.

    a. In this and earlier Contra Costa studies mutagens (and PAH) were significantly correlated with fine fraction Pb and Br, indicating contributions from primary automotive emissions.

    b. Chemical analysis by other investigators has identified mutagens (various PAH and nitroarenes) in on-road vehicle particulate emissions (20) as well as other combustion source particulate matter (21).

c. Studies of upwind-downwind freeway data in Los Angeles by Sweetman et al (22) have demonstrated an incremental burden of direct mutagens in aerosol attributable to freeway traffic which was comparable to the area wide background mutagen density.

2. Many results suggest that some mutagens behaved as secondary aerosols. The hypothesis that some mutagenic aerosols are formed in the atmosphere is strengthened by the following evidence:

a. During pollution episodes in Contra Costa County, mutagens were positively correlated with NO^-_3, assumed to be a secondary aerosol tracer. The association of mutagenicity with NO_3^- occurred area-wide.

b. Pitts and co-workers (23) observed that ratios of mutagen densities (rev/m^3) to CO were generally higher at Riverside, a receptor site, than at El Monte, an intermediate receptor location in the Los Angeles basin. Since CO is an unreactive combustion emission, the mutagen density/CO ratio takes into account variations in emissions and atmospheric dispersion. Higher ratios at Riverside suggest atmospheric mutagen formation during aerosol transport from Los Angeles.

c. The ratios of mutagenic densities to Pb which we have measured in Contra Costa County in this and a previous study (18) were highest during summer episodes when the prevailing atmospheric conditions (i.e. hot, dry, stagnant) favored chemical transformations. Since Pb, like CO, is an unreactive emission, the mutagenic density/Pb ratio should also take into account variations in (automotive) emission profiles and dispersion. Thus the high ratios during episodes in August 1981 (18) and September 1983 (shown below) may reflect atmospheric mutagen formation.

d. Smog chamber studies have demonstrated the formation of nitro-PAH and other mutagens. Mutagenicity of some nitro-PAH's exceed

the mutagenicity of the parent PAH by several orders of magnitude in laboratory analysis. Some of these highly mutagenic nitro-PAH are known to be primary pollutants emitted by various combustion sources. However chamber studies (24,25) have also shown that irradiation of mixtures of atmospheric hydrocarbons, nitric acid (HNO_3) and reactive gases (NO_2, O_3) can lead to mutagen formation. Thus some some hydrocarbons may be converted to secondary mutagenic products under simulated atmospheric conditions.

e. Measurements in a nitroreductase mutant indicate the likeley presence of nitroorganic mutagens. Less than 10% of the total mutagenicity in ambient air samples is due to identified PAH. Thus most of the mutagenicity remains to be explained in chemical terms. A substantial proportion of this excess mutagenicity may be due to highly mutagenic nitroarenes, which are not only ubiquitious primary pollutants but may also be derived from secondary atmospheric transformations. We infer that nitroarenes were probably major contributors to the mutagenicity of Contra Costa aerosols from the fact that mutagenic activities of aerosol extracts were two to three times lower in a Salmonella strain (TA98NR) deficient in an enzyme for some mononitroarene activation, than in the standard tester strain (TA98). With respect to mutagenicity of community air collected in other cities this finding is not unique. For example, air particulate samples from Los Angeles (23) and Detroit (26) also showed markedly reduced mutagenic activities in nitroreductase deficient strains.

f. Finally, measurement artifacts confound the secondary mutagen hypothesis. The positive correlations of mutagenic density with NO_3^- and the demonstration that mutagenic organic compounds can be formed under simulated atmospheric conditions support the hypothesis of secondary formation of mutagenic aerosols in the atmosphere. However interpretation is complicated by measurement artifacts in nitrates and nitro-aromatic compounds. The association between mutagens and NO_3 could be influenced by HNO_3 artifacts

produced by sampling on glass fiber filters. There are two concerns. Gas phase HNO_3 can bind to glass fiber and artificially increase apparent particulate NO_3^- concentrations (27). More importantly, gas phase HNO_3 may catalyze chemical transformations of PAH to produce highly mutagenic nitroaromatic compounds during sample collection on glass fiber (13). The significance of these potential artifacts can not be assessed accurately at present.

3. For the first time, industrial contributions to mutagenic aerosols were also suggested by significant positive correlations between mutagenic density and S (both fine fraction S and SO_2) at Richmond and Martinez. These sulfur oxides are major air pollutants in the vicinity of large oil refineries and chemical plants concentrated in Contra Costa County. The major industrial sources are refineries in Richmond (Chevron), Martinez (Shell, Tosco) and Benicia (Exxon) and a chemical plant in Rodeo (Union) (28).

Routine collection and analysis of seasonal composite filters in Contra Costa County between 1979-1984 (Chapter IV) revealed both seasonal variations and trends.

1. Concentrations of mutagens, PAH and the standard air pollutants (TSP, Pb, NO_3^-, $SO_4^=$) were highest during the winter (Nov.-Feb.) season. PAH exhibited the greatest seasonal changes, 3-10 fold. High wintertime PAH concentrations could reflect contributions from residential wood combustion.

2. A positive trend in concentrations of mutagenic aerosols ($\pm$S9) was found between 1979 and 1984. For example, a nearly four-fold increase in the annual average mutagenic density (+S9) from 5 to 19 rev/m^3 was observed over the five years of monitoring.

3. The positive trend in mutagenicity was in contrast to the fairly constant (annual average) levels of PAH and the decreasing levels of the standard pollutants. The decrease in Pb was most apparent. For example over

the five winter seasons (1979-1984) Pb decreased from 0.57 ± 0.13 ng/m^3 to 0.27 ± 0.03 ng/m^3. The Pb gasoline phase-out program in the Bay Area, or different meteorological factors for the sampling seasons may be responsible.

E. Recommendations for Future Research

The partial answers derived from the present research effort also generated additional questions for possible future research.

Investigation of sources has lead to the suggestion that mutagens may be formed atmospherically, during normal aging of community aerosols. Before endorsing this suggestion further, several measurement questions must be addressed. As noted above, the apparent association between mutagens and NO_3^- could be influenced by HNO_3 artifacts produced by sampling on glass fiber filters. Gas phase HNO_3 can bind to glass fiber and artificially increase apparent particulate NO_3^- concentrations. Appel and co-workers (27) have recently compared artifact NO_3^- formation on different filter media. Laboratory and atmospheric sampling studies were performed to evaluate glass fiber and Teflon filters for their abilities to form artifact particulate nitrate with HNO_3. At nitric acid dosages representative of those in the atmosphere, glass fiber filters retained >94% of the HNO_3 and Teflon <2% of HNO_3.

Gas phase HNO_3 may also catalyze chemical transformations of PAH to produce highly mutagenic nitroaromatic compounds. These transformations can occur both in the atmosphere and on filters during sample collection. Pitts et al (13) first showed the formation of directly mutagenic nitroderivatives from PAH coated on glass fiber filters and exposed to flows of air containing NO_2 and traces of nitric acid. Extending this research, Pitts and co-workers (23) have more recently studied sampling artifacts utilizing two filter types (glass fiber and Teflon-impregnated glass fiber). The ratios of mutagen densities for POM simultaneously collected on glass fiber and Teflon-impregnated glass fiber varied by more than a factor of ten. The greatest differences occurred during periods of elevated O_3 concentrations, suggesting that under such conditions there is an artifact effect associated with particulate collection (probably) on glass fiber

filters. Ambient concentrations of HNO_3 and other reactive gases (NO_x, O_3) in Contra Costa County are not as high as in El Monte and Riverside, where these artifacts were studied. Nevertheless, direct evaluation of possible HNO_3-glass fiber effects in Contra Costa air samples should be done. Experiments are recommended to compare mutagenicity and NO_3^- values in aerosols collected on glass-fiber and Teflon-impregnated glass fiber filters in samplers equipped with or without HNO_3 denuders.

A further recommendation concerns industrial emissions. We have observed for the first time in Contra Costa County significant positive correlations between mutagenicity and the petrochemical tracer S at Richmond and Martinez. Petrochemical and other chemical sources may therefore contribute to mutagenic emissions. Follow-up research on stationary source emissions should be done. This research should provide sampling methods for both volatile and aerosol mutagens; at Richmond and Martinez mutagenicity was positively correlated with gaseous SO_2, as well as fine S aerosols.

A final recommendation is to maintain and expand the monitoring network for mutagens and PAH, in light of the increasing trends in mutagenicity observed in recent years. To verify the trend analysis, routine monitoring should continue in Contra Costa County and be extended to include other high pollution locales in the Bay Area (e.g. southern Santa Clara County) and adjacent air basins (e.g. Sacramento-San Joaquin Valley, Chico to Bakersfield). Existing air sampling networks would be used. Because samples are routinely collected at sites in these networks, and Ames and PAH testing are routinely carried out in AIHL, the cost would be minimal.

CHAPTER II.

APPLICATION OF A SALMONELLA MICROSUSPENSION PROCEDURE TO THE MEASUREMENT OF MUTAGENICITY IN AIR PARTICULATE MATTER: HIGH RESOLUTION DIURNAL VARIATIONS

A. Summary

A simple modification of the Salmonella liquid incubation assay (19) was used to determine mutagenic activity of airborne particulate matter. The modification consists of adding ten times more bacteria (approximately 10^9 per incubation tube) and five to ten times less metabolic enzymes compared to the plate incorporation method. The mixture volume is approximately 0.2 ml and the mixture is incubated for 90 minutes before pouring it according to the standard protocol. The modified procedure was approximately 10 times more sensitive than the standard plate incorporation test for detecting mutagens in air particle extracts and approximately 13-30 times more sensitive for the chemical mutagens 2-nitrofluorene, 4-nitroquinoline-N-oxide, 2-aminofluorene and benzo(a)pyrene in bacterial strain TA98. This microsuspension procedure was applied to air particulate samples collected with low volume (15-50 liters per min) virtual dichotomous air samplers. Mutagenic activity was detected in particle extracts obtained from one cubic meter of air or less (17 µg of extract) and was associated exclusively with fine particles (aerodynamic diameters of less than 2.5 µm). Diurnal patterns of mutagenic activity (TA98 revertants per cubic meter air) were investigated by measuring filter extracts from two-hour samples collected in three San Francisco Bay Area cities during air pollution episodes. Four criteria pollutants - lead, nitrogen dioxide, ozone and sulfur dioxide were simultaneously sampled at one location. Mutagenicity from fine particles sampled at this location was highly correlated with lead and much less correlated with nitrogen dioxide, ozone and sulfur dioxide. The microsuspension procedure is applicable in testing samples of limited mass.

B. Introduction

Mutagenic activity of solvent extracts from community air particulate matter has been studied by a number of investigators (1-6). The activity is a rough index of exposure to potential carcinogens, aids in the chemical characterization and identification of mutagens and helps better define the sources of chemical mutagens. The Salmonella typhimurium/microsome test (12) has often been used in air pollution mutagen studies. It is the most validated of the short-term genotoxicity tests and is convenient and economical to use. The airborne particulate matter used in mutagenicity studies are collected by samplers usually of the hi-vol, cascade or electrostatic precipitator type, which draw large volumes of air through filters to provide enough sample mass for subsequent biological or chemical testing. Hi-volume-type samplers have also been combined and operated simultaneously (29) to acquire several times as much material as a single hi-vol sampler. Limited numbers of certain hi-volume samplers are available and for some of them, such as the ultra high volume sampler (17), mobile deployment is difficult due to the large size of the instrument. Furthermore, the more volatile mutagens adsorbed onto the particles may be lost or chemically transformed because such a large volume of air passes over the particle sample (30).

The problems of sampling can be reduced by the use of more sensitive bioassays to detect mutagenicity in samples of limited mass. The more sensitive assays would also facilitate subsequent separation and identification of specific mutagens.

We report here progress in using a highly sensitive modification of the Salmonella liquid incubation assay to measure the mutagenicity of airborne particle extracts. The simple modification was previously described for detecting mutagens in cigarettes smokers' urine (19) with an increase in sensitivity of approximately 20 times that of the plate incorporation test. We describe first, the relative sensitivity of the modification to the plate incorporation test using known mutagens and second, the initial application of the modification for measurement of mutagenic activity in a composite air filter extract and filter extracts taken from low volume, size selective dichotomous samplers.

C. Materials and Methods

1. Chemicals

Benzo(a) pyrene (BP, Gold Label), 2-aminofluorene (2-AF), 2-nitrofluorene (2-NF) and 4-nitroquinoline-N-oxide (4-NQO) were purchased from Aldrich Chemical company, Milwaukee, Wisconsin and were used without further purification. The extraction solvents (methanol, dichloromethane and toluene) were glass-distilled OmniSorb brand purchased from Matheson, Coleman and Bell, Gibbstown, New Jersey. Dimethyl sulfoxide was Photo-rex grade and was purchased from J.T. Baker Chemical Company, Phillipsburg, New Jersey.

2. Criteria Gas Pollutant Sampling and Analysis

At one sampling site (Martinez, California), gaseous air pollutants were simultaneously measured by the Bay Area Air Quality Management District, using a mobile sampling van. Ozone was measured by ultraviolet absorption with a Dasibi model 1003-AH Ozone Monitor; Nitrogen dioxide was measured by chemiluminescence with a Thermal-electron Model 14D analyzer and Sulfur dioxide was measured by fluorescence using a Thermal-electron Model 43 pulse-fluorescence analyzer. All these methods are EPA reference methods or have been certified as equivalent (31).

3. Air Particle Collection and Sample Preparation

The plate incorporation and the microsuspension procedures were compared using a composite filter extract from 24-hour hi-vol samples collected for 10 consecutive days during the summer of 1982. Particulate samples were collected on 8 x 10 inch glass-fiber filters (EPA equivalent from Whatman Ltd., Springfield Kent, England.) The hi-vol sampler had a flow rate of 1 m^3/min and was placed on the roof (approximately 30 meters above street level) of the Department of Health Services Building, Berkeley, California.

Collections of size-segregated "fine" (<2.5 μm aerodynamic diameter) and "coarse" (2.5-15 μm aerodynamic diameter) air particulate fractions were made at Rodeo, California during the summer of 1982 and at Berkeley and Martinez, California, during the fall of 1982 using dichotomous air samplers. The town of Rodeo is located approximately 10 miles north of Berkeley. A major freeway and chemical plants are nearby. At Rodeo, size-segregated samples were collected with a standard Sierra Model Dichotomous sampler (Sierra Instrument Corp., Carmel Valley, CA) operated at a flow rate of 16.7 liters/min (l/min). Teflon filters (37 mm diameter and 2 μm pore size were purchased from Membrana, Inc., Pleasanton, CA and were changed manually every 2 hours for a total collection period of 24 hours. At Berkeley and Martinez, air samples were collected using an automatic dichotomous sampler (32) provided by the Lawrence Berkeley Laboratory (LBL), Berkeley, CA. Filters were 37 mm diameter, 1 μm pore size, and came mounted on plastic frames (Membrana, Inc., Pleasanton, CA). The sampling flow rate was 50 liters/min.

Dichotomous filters were extracted by sonication in a mixture of 1:1:1 methanol, dicholoromethane and toluene (trisolvent) as previously described (33). Filters were extracted in 16 x 125 mm screw-top glass tubes; 4 ml of extraction solvent was added to each tube, which was then sealed with a Teflon-lined screw cap and placed in an ultrasonic water bath at 45°C. After sonication at maximum power for 20 minutes, the extract was passed through a 0.5 μm Fluoropore filter. The filter was washed again with 3 ml trisolvent by sonication, the extract filtered and combined with the initial filter extract. The volume of the combined extract was decreased tenfold in vacuo by rotary evaporation at 45°C, and the extract was transferred to a 1 dram vial, evaporated under a stream of nitrogen to dryness, capped under nitrogen and stored at -20°C until tested. All extraction procedures were carried out under yellow fluorescent lights to minimize potential photooxidation.

Lead in dichotomous filter samples was determined by atomic absorption spectrophotometry (34). A sample 10 mm in diameter from the center

of the filter was extracted in 10% nitric acid, and the extract analyzed for lead with a Perkin-Elmer Model 503 Atomic Absorption Spectrometer.

4. Mutagenicity Assays

All mutagenicity testing was done using frame shift tester strain TA98 (35) and nitroreductase deficient derivative, TA98NR (36). The standard plate incorporation method for detecting mutagens with the Salmonella/mammalian microsome test was performed as described by Ames et al. (12). A liver extract prepared from male Spraque Dawley rats (150-200g) treated with Aroclor 1254 was prepared according to the method of Ames et al. (12). The protein concentration was 30 mg/mL, determined by the method of Lowry et al. (37). A simple modification of the Salmonella liquid incubation procedure reported by Kado et al. (19) was used throughout.

Single colonies were taken from a master plate made from Oxoid Nutrient Broth (Oxoid Ltd., Hants, England), added to 10 ml of Oxoid Nutrient broth and grown overnight to a concentration of approximately 1-2 x 10^9 cells per ml. Cells were concentrated by centrifugation (10,000 X g, 4^oC), 10 minutes and resuspended in ice-cold phosphate buffered saline (PBS, 0.15M, pH 7.4) to a concentration of 1 X 10^{10} cells per milliliter. The microsuspension procedure was performed with metabolic activation (+S9) by adding the following ingredients, in order, to 12 X 75 mm sterile glass culture tubes placed in ice: 0.1 ml S9 mix, 0.005 ml of DMSO solution containing the test material and 0.1 ml of concentrated bacteria (approximately 1 X 10^{10} per ml PBS or 1 X 10^9 per tube). A similar mixture was prepared to test samples without the addition of metabolic enzymes (-S9), except that the sample (in DMSO) was added to the concentrated bacterial solution first, followed by the addition of 0.1 ml phosphate buffer (0.1M, pH 7.4). The tubes were capped and incubated in the dark at 37^oC with rapid shaking. After 90 minutes, the tubes were placed in an ice water bath, removed singly from the ice bath and 2 ml of molten top agar containing 90 nmoles of both histidine and biotin were added. The molten suspensions were immediately mixed with a

Vortex mixer and poured into minimal glucose plates. Plates were incubated at 37°C in the dark for 48 hours and were counted using an automatic colony counter (Biotran III, New Brunswick Scientific, Edison, N.J.). Genetic markers for the strains were routinely verified. Mutagenicity testing was carried out in a room fitted with yellow fluorescent lights to minimize potential photooxidation.

Duplicate aliquots of all mutagen standards and extracts of air particulate matter were tested at 3 or more doses.

D. Results and Discussion

1. Chemical Mutagens

Mutagenic activities of the chemical mutagens: 2-nitrofluorene (2-NF), 4-nitroquinoline-N-oxide (4-NQO), 2-aminofluorene (2-AF) and benzo(a)-pyrene (BaP) were determined by the standard plate incorporation assay and the microsuspension procedure. The microsuspension procedure measured much higher levels of specific mutagenic activity for each chemical; the activity of 2-NF increased most dramatically, by a factor greater than 30 (Table II-1). There was little increase in the number of spontaneous revertants in the microsuspension procedure although ten times more bacterial cells were added. For example, the solvent blanks in TA98 for the microsuspension and standard Ames assays (-S9) were 29 and 17 revertants per plate, respectively. This can be explained as follows: The number of spontaneous revertants is related to the total number of cell divisions which occur during 48 hours of incubation. In both assays approximately the same total number of divisions occur, because growth is limited to the same extent by the available histidine. Since ten times more cells are added initially in the microsuspension procedure, fewer divisions per cell take place by the time the final (histidine-limited) cell density is reached. However, in the plate incorporation test there are initially fewer cells added per plate, but more divisions per cell. Thus the total number of divisions and therefore the number of spontaneous revertants which occur in both procedures are similar.

TABLE II-1

COMPARATIVE MUTAGENIC ACTIVITY OF MUTAGENS IN THE PLATE INCORPORATION AND MICROSUSPENSION PROCEDURES

Chemical	Specific Mutagenic Activity[a] (TA98 rev/nmol) Plate Incorporation	Micro-Suspension	Fold Increase in Sensitivity
Benzo(a)pyrene	93	907	10
2-Aminofluorene	199	2460	13
2-Nitrofluorene	61	1940	31
4-Nitroquinoline-N-oxide	103	1800	18

[a]Determined from the linear portion of the dose-response curve from a single experiment.

The direct-acting mutagens 2-NF and 4-NQO were 20-30 times more mutagenic in the microsuspension procedure than in the plate incorporation assay and the indirect-acting mutagens BaP and 2AF were approximately 10 times more mutagenic. The results for BaP are in good agreement with the previous study (19) where the microsuspension procedure was about 14 times more sensitive. We also investigated the applicability of the microsuspension procedure to a related tester strain, TA98NR. As shown in Table II-2, the mutagenic activity of 2-NF decreased appreciably when it was tested in TA98NR, but the activity of 4-NQO remained approximately the same. These responses are similar to those reported by Rosenkranz and Mermelstein (38) for the plate incorporation test. The mutagenic activity of the pooled air extract also decreased from 24 rev/m^3 to approximately 4 rev/m^3, indicating that compounds similar to 2-NF may be responsible for most of the "direct-acting" mutagenic-activity in this sample. The increased sensitivity of the microsuspension procedure for both direct and indirect-acting mutagens is probably due to the combined effects of increasing the total number of bacteria added and concentrating the incubation mixture including the sample in a small volume (0.2 ml). The former increases the concentration of bacterial DNA targets available for interaction with mutagens, and the latter increases the likelihood of mutagens being taken up by the cells.

2. Hi-vol Air Particle Extracts

Dose response curves for mutagenic activity of the composite hi-vol air particle extract, constructed from the plate incorporation test and from the microsuspension procedure are illustrated in Figure II-1. The amount of extract added is expressed in units of "cubic meter equivalents", the number of cubic meters of sampled air containing a specific amount of particulate matter. One cubic meter equivalent (m^3 equivalent) is approximately equal to 17 µg of particulate matter for the composite sample. The extract added per plate in the microsuspension procedure and plate incorporation test respectively, was 1-11 m^3 equivalents (23-185 mg of particulate matter) and 5-43 m^3 equivalents (92-739 mg of particulate matter). The optimal levels of S9, determined to be 600 µg protein/plate

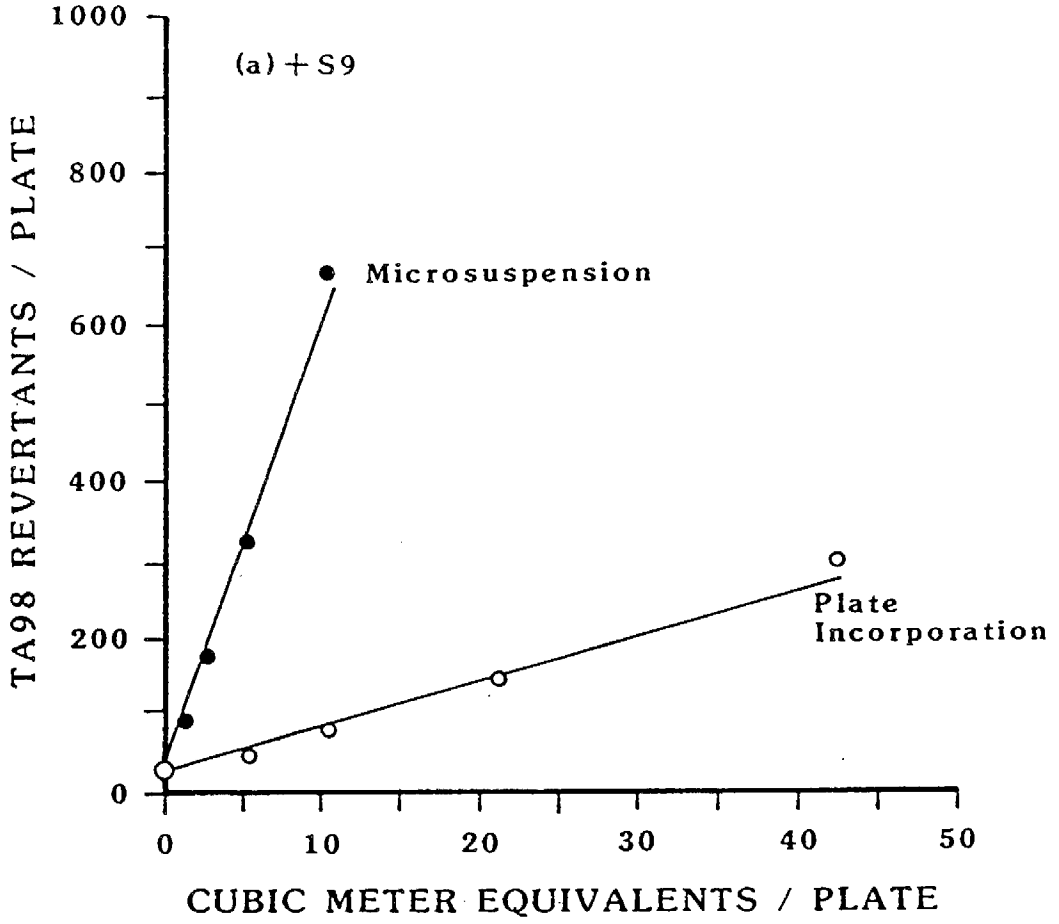
TABLE II-2

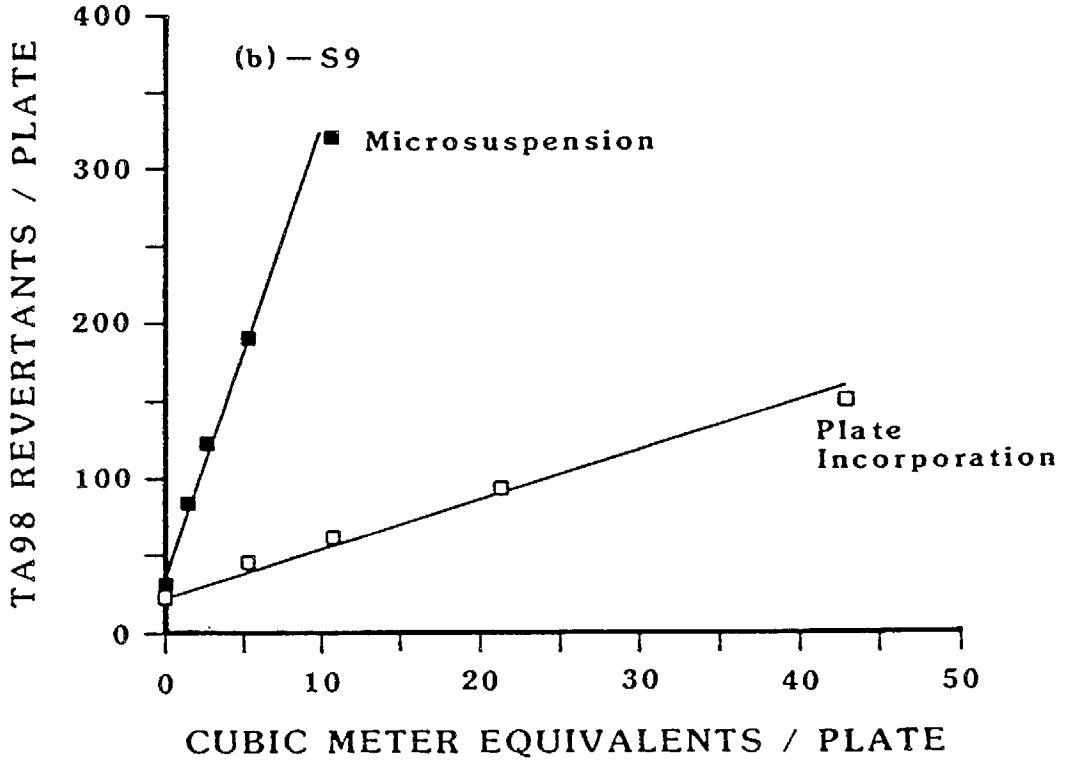
COMPARISON OF DIRECT MUTAGENIC ACTIVITY OF 2-NITROFLUORENE, 4-NITROQUINOLINE-N-OXIDE AND COMPOSITE BERKELEY AIR FILTER EXTRACT IN TA98 AND TA98NR AS DETERMINED BY THE MICROSUSPENSION PROCEDURE

Test Substance	Specific Mutagenic Activity[a] TA98	TA98NR
2-Nitrofluorene (rev/nmol)	4170	405
4-Nitroquinoline-N-oxide (rev/nmol)	1540	1180
Composite Berkeley Air Filter Extract (rev/m^3)	24	4

[a]Calculated from dose-response curve using pooled data from 2 experiments.

FIGURE II- 1. Dose-response curves for composite hi-vol air particle extract. Determined using the plate incorporation test and microsuspension procedure with (a) and without (b) rat liver S9.





for the standard plate incorporation test and 30 µg protein/plate for the microsuspension procedure, were used for all the tests. As illustrated in Figure II-1, the microsuspension procedure was approximately 10 times more sensitive than the plate incorporation test, both with and without metabolic activation. The respective slopes for the microsuspension procedure with and without S9 were approximately 60 and 26 rev/m^3, while the corresponding slopes for the plate incorporation test were 6 and 3 rev/m^3. A response was considered positive if it was at least twice the number of spontaneous revertants. The microsuspension procedure and the plate incorporation assay required air samples of approximately 1 m^3 and 10 m^3, respectively, to achieve this doubling. The concentrations of total suspended particulates in the air samples used to prepare the composite were between 50-100 µg/m^3. The amount of S9 protein required per plate in the microsuspension procedure was one-twentieth that needed in the plate incorporation test. These results are consistent with those obtained during the analysis of urine from cigarette smokers reported previously (19).

3. Diurnal Variations in Mutagenicity of Fine Particle Extracts

Data on diurnal variations in mutagenicity were obtained from two-hour samples collected by dichotomous samplers. The first of the three diurnal studies was done in Rodeo, California. Two-hour samples were collected during the 24 hours beginning at 6 a.m., August 27, 1982, using a Sierra manual dichotomous sampler at a flow rate of 16.7 liters per minute. Filters were changed manually every 2 hours. As illustrated in Figure II-2, mutagenic activity was detected with metabolic activation (+S9) in extracts of the fine fraction (<2.5 µm), and a distinct diurnal pattern of mutagenicity can be seen, with a morning peak of activity between 10 a.m. and 12 noon and an evening peak between 8 and 10 p.m. In this experiment, activity was not detected in the fine fraction extracts in the absence of S9 and none was detected in the coarse fraction extracts, whether or not S9 was present. The diurnal variations in mutagenic activity in the Rodeo extracts, although not especially large, encouraged us to carry out a second study under circumstances where higher activities were anticipated.

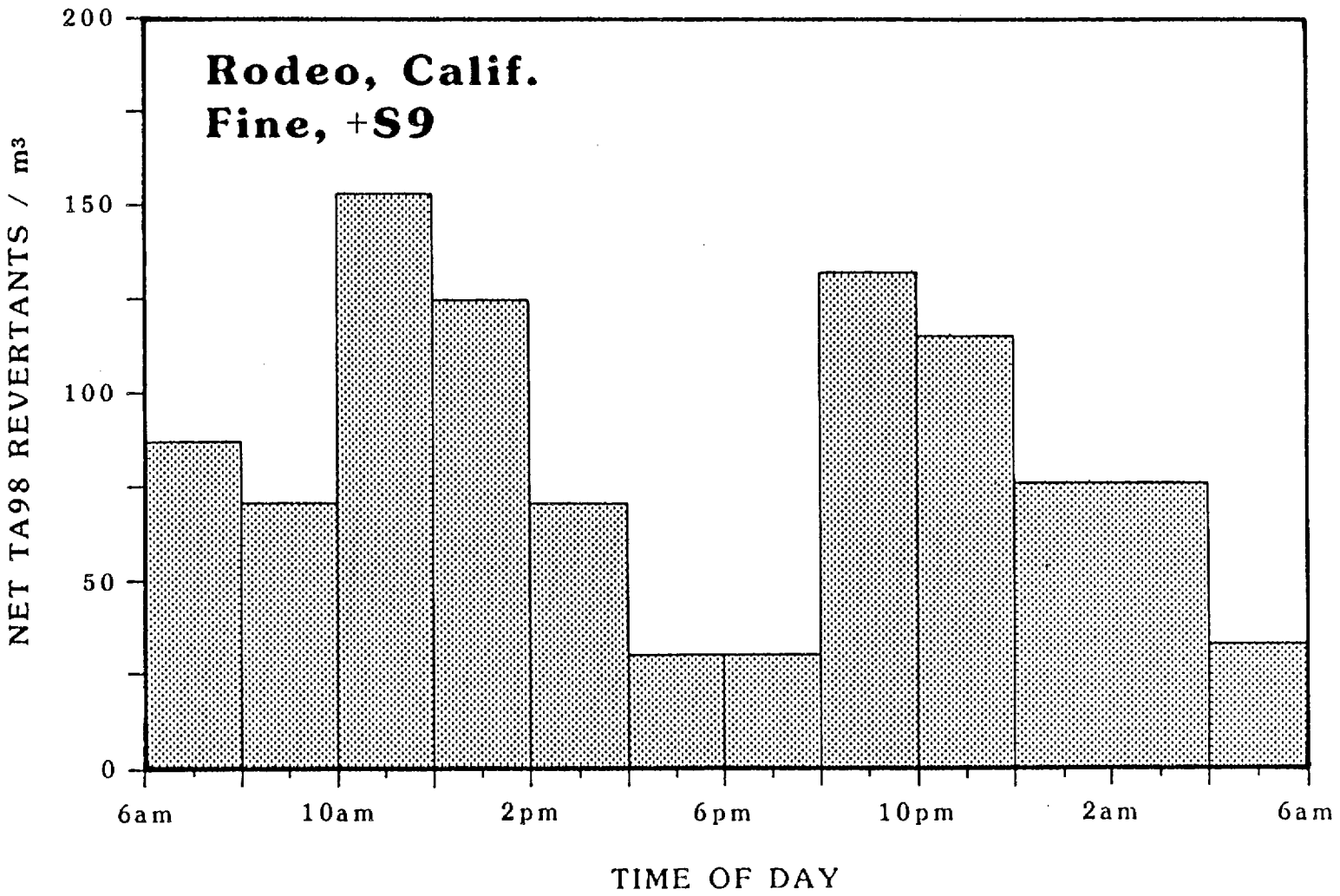


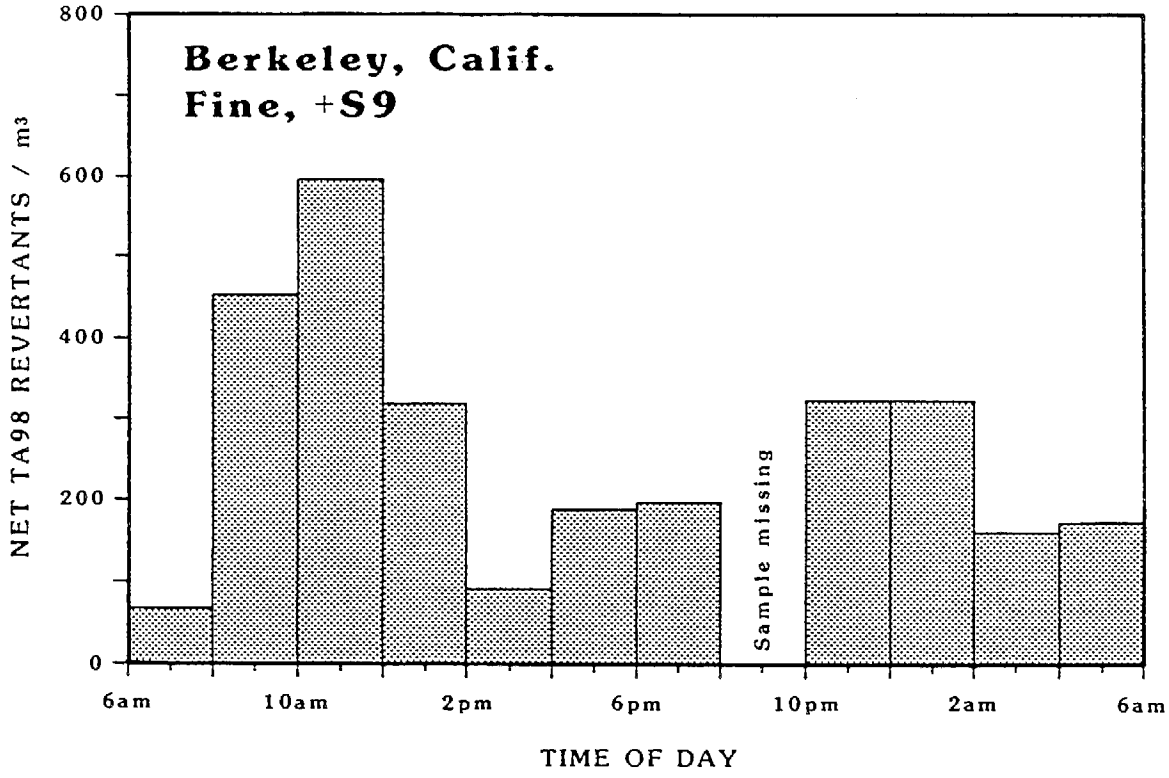
FIGURE II- 2. Diurnal variation of mutagenicity of fine airborne particles collected in Rodeo, California and measured in the microsuspension procedure. A single four hour sample was collected between midnight and 4 a.m.

The second experiment was conducted in Berkeley on October 20-21, 1982. Two-hour samples of size-segregated air particles were collected with an automatic dichotomous sampler (ADS/LBL Model I) located in a service yard outside the California Department of Health Building in downtown Berkeley and operated at a flow rate of 50 l/min. The diurnal variations observed are depicted in Figure II-3. Mutagenic activity of fine particle extracts from the samples ranged from less than 75 to nearly 600 revertants per cubic meter of air sampled. Similar diurnal patterns of mutagenic activity were detected both with and without metabolic activation, the +S9 response being approximately three times higher. Virtually no activity was detected in the coarse particle extracts. The maximum level of activity measured in Berkeley was about four times higher than that measured in Rodeo, and the diurnal patterns were similar at both locations. A morning mutagenicity peak occurred between 10 a.m. and 12 noon and an evening peak between 10 p.m. and 2 a.m. Both the morning and evening peaks appeared later than peak traffic flow (7-8 a.m. and 5-6 p.m.). The success of these first two trials prompted a third field study in which levels of mutagenicity and criteria air pollutants were measured simultaneously to better define the possible sources of activity.

Air sampling for a third field trial was conducted at the Mountain View Sanitary District's facility in Martinez, California during the 36 hours beginning at 6 p.m., November 3, 1982. The criteria air pollutants samples were lead (Pb), nitrogen dioxide (NO_2), ozone (O_3) and sulfur dioxide (SO_2). The two-hour particulate samples analyzed for mutagenicity and lead were collected with the LBL Model 1 Automatic Dichotomous Sampler (ADS); only the fine fractions were analyzed. Gaseous pollutants were monitored continuously and hourly averages were calculated and used for comparisons. Twelve-hour hi-vol samples were collected simultaneously at the site.

The results are illustrated in Figure II-4. Peak levels of mutagenic activity both with and without S9 were found in the early morning around 6 a.m. and around midnight. Maximum values, measured in the presence of metabolic activation, were greater than 1,000 revertants/m^3 air.

FIGURE II- 3. Diurnal variation of mutagenicity of fine airborne particles collected in Berkeley, California and measured in the micro-suspension procedure with (a) and without (b) addition of rat liver S9.



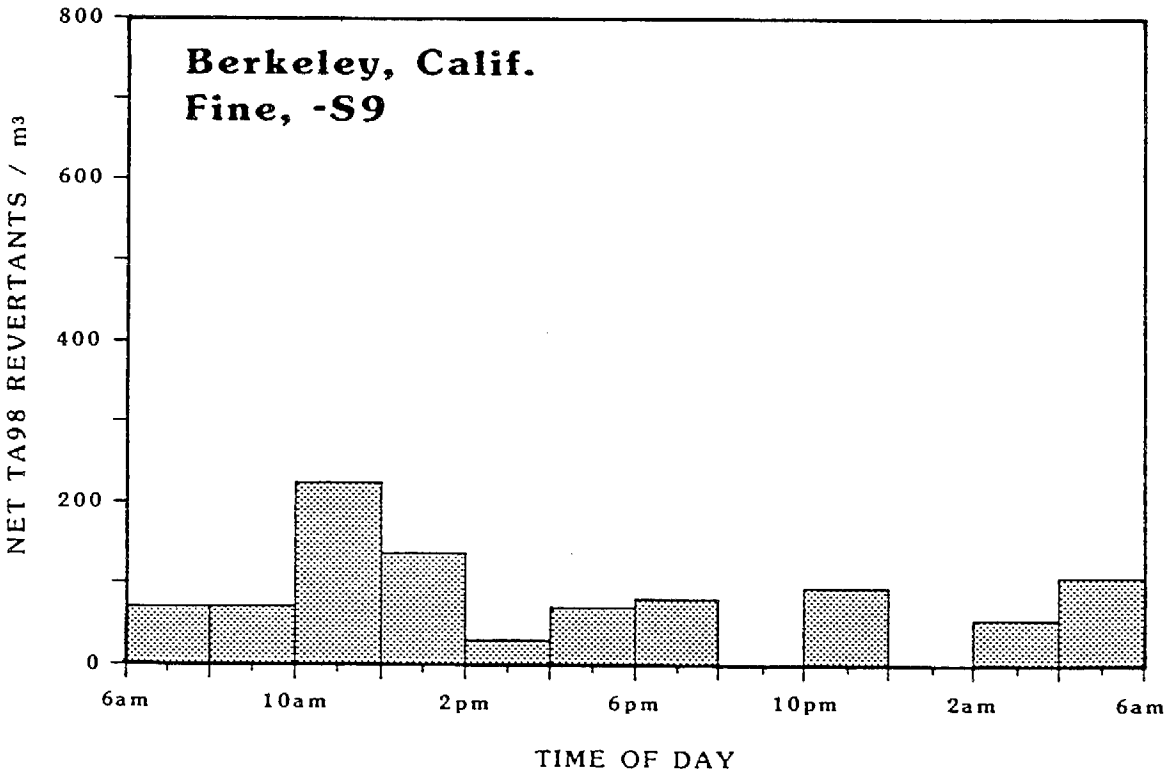
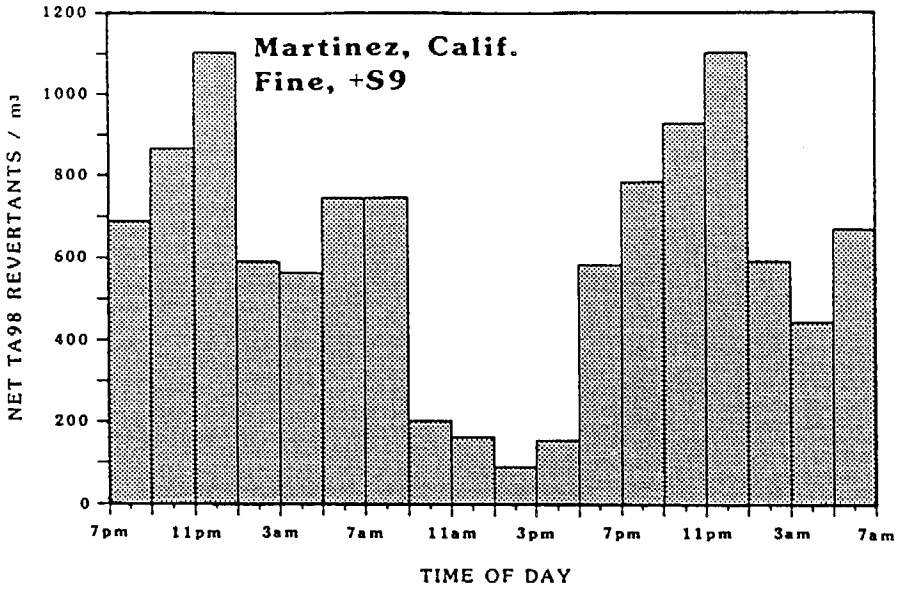
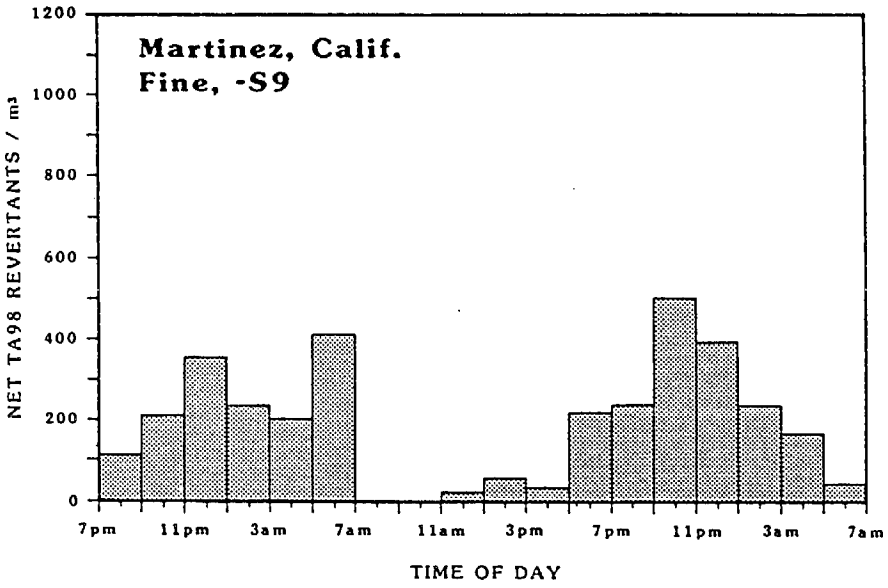
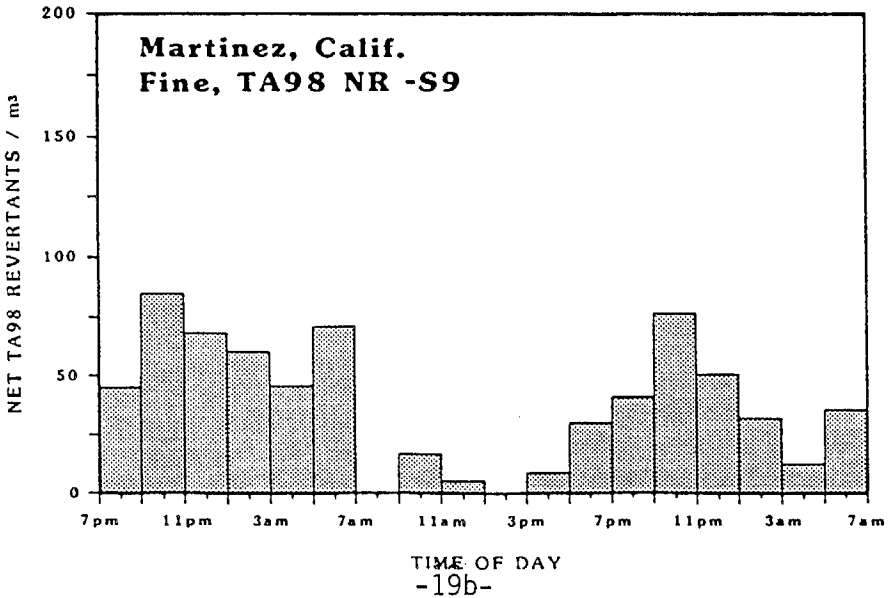


FIGURE II- 4. Diurnal variation of mutagenicity of fine airborne particles collected in Martinez, California and measured in the microsuspension procudure. TA98 with S9 (a); TA98 without S9 (b); TA98 NR without S9 (c).







Mutagenic activities of the hi-vol samples taken in parallel with the dichots were compared to the calculated average activities of the dichots. As summarized in Table II-3, the calculated average activities of the dichotomous samples are similar to the activities of the hi-vol sample. Although the average mutagenic activity of the dichot is slightly higher for the two nighttime periods, especially for mutagenic activity dependent on metabolic activation these differences are within experimental error.

The diurnal pattern of fine fraction lead (not shown) was very similar to that of mutagenicity, exhibiting both early morning and late night peaks. Lead and mutagenicity are strongly correlated ($r = .92$), as shown by the plot of sample values in Figure II-5. Since motor vehicles are the primary source of airborne lead, this correlation suggests that they are also the source of much of the airborne mutagenic activity.

Diurnal patterns of the three measured gases (O_3, SO_2, NO_2) did not correlate well with mutagenic activity. Only lead concentrations were related to concentrations of particulate mutagenicity.

The present results may be compared with those of Pitts and coworkers (22,29). In their studies, diurnal comparisons were made of airborne mutagencity of Los Angeles air using 3-hour hi-volume samples. They found that mutagenic activity was strongly correlated with carbon monoxide (CO), emitted principally from automobile emissions in Los Angeles air, and that mutagenic peaks were closely related to peak commuting hours. In the present study, mutagenic peaks appeared later than would be expected from diurnal patterns of traffic flow near the sampling sites.

Our conclusion that mobile source emissions contribute significantly to the mutagenicity of airborne particles sampled in Martinez is consistent with results of a related study which investigated sources of particulate matter collected at four Contra Costa County locations during seasonal pollution episodes in 1981-82 (18,39). Air samples were analyzed for

TABLE II-3

MUTAGENICITY OF PARTICLES COLLECTED BY HI-VOLUME AND DICHOTOMOUS AIR SAMPLERS RUN IN PARALLEL AT MARTINEZ, CALIFORNIA

	Mutagenic Activity (TA98 rev/m^3)			
	+S9		-S9	
Sampling Period	Hi-Vol[a]	Dichot[b] (Ave)	Hi-Vol[a]	Dichot[b] (Ave)
19:20-7:05 (11/3-11/4/82)	572	723	223	238
7:10-19:15 (11/4/82)	304	236	101	86
20:20-7:05 (11/4/-11/5/82)	624	727	238	296

[a]Mutagenic activity determined from linear portion of dose-response curve.

[b]Mutagenic activity is the average number of revertants per cubic meter for the 12 hour sampling period calculated from six consecutive 2-hour sampling periods.

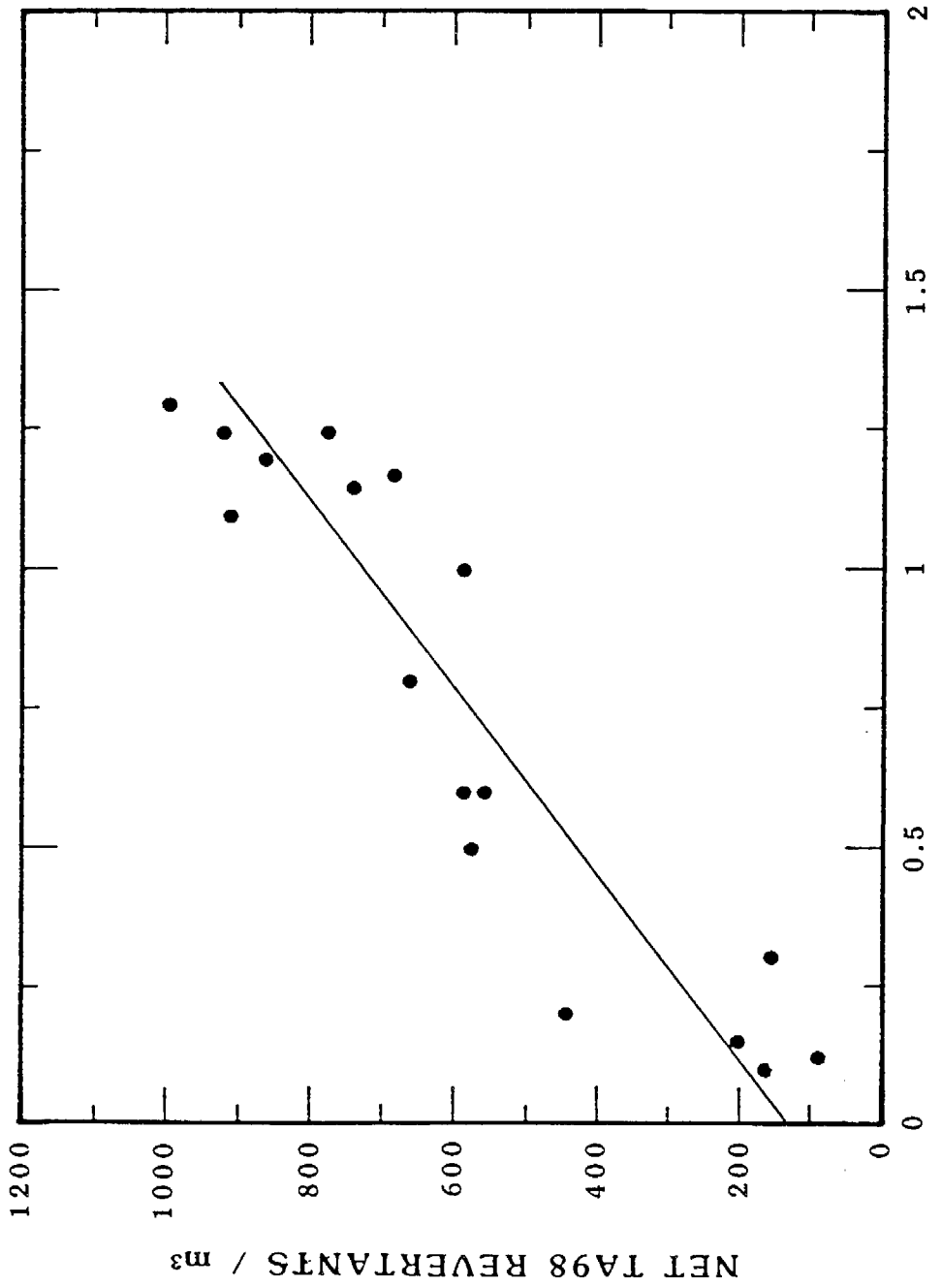


FIGURE II- 5. Correlation of airborne lead and mutagenicity (microsuspension procedure with S9) from fine particles collected at Martinez, California sampling site. r = 0.92.

mutagenic activity and a variety of particulate chemical pollutants and gases. Mutagenicity was found to be strongly associated with lead-containing fine particles.

The present study is also in agreement with previous studies on size-segregated particles, in which investigators found that most of the mutagenic activity is associated with particles of diameters of about 2 μm or less (40,41).

E. Conclusions

This study presents data on diurnal variations in mutagenicity of community aerosols of less than 2.5 μm aerodynamic diameter in samples of 2 hour duration. In field studies, diurnal variations in mutagenic activity (revertants/m^3) of 10 fold were found. Variations in mutagenic activity correlated well with the variations in fine-fraction lead, implicating motor vehicles as a significant source of mutagens. These experiments were made possible by the use of the highly sensitive microsuspension modification of the Salmonella liquid incubation assay. This modification makes possible high resolution diurnal studies of fine aerosols and can be applied in future studies where sample mass is a limiting factor.

CHAPTER III

SOURCES OF MUTAGENS AND POLYCYCLIC AROMATIC HYDROCARBONS IN CONTRA COSTA COMMUNITY AEROSOLS DURING POLLUTION EPISODES: DIURNAL, GEOGRAPHIC AND EPISODE VARIATIONS

A. Introduction

As described previously, applications of the Ames Salmonella test (12) to community air particles have demonstrated that chemical mutagens are ubiquitous components of urban aerosols (1-6). A fundamental problem concerns source identification. The measure of a relatively high mutagenic activity in a given geographical area is of limited value unless the sources of the mutagenicity can be identified and therefore potentially controlled. In a previous CARB-supported air pollution study in Contra Costa County, AIHL measured mutagenicity and a variety of chemical air pollutants (18). The study examined diurnal variations of mutagenic activity and the relationship of mutagenic activity to other aerosol variables, including certain source tracer elements. The results indicated that mobile sources were significant contributors to PAH and particulate mutagens. The present study extends this earlier research using the same experimental approach.

B. Experimental Methods

1. Air Sampling and Site Descriptions

Six 36 hour sampling episodes were carried out in Contra Costa County during periods of high pollution in 1982-1984. Samples were collected at four locations in Richmond, Martinez, Concord and Pittsburg (Figure I-2). Three (Richmond, Concord and Pittsburg) are located so as to reflect the quality of outdoor community air breathed by the public. These three are permanent stations of the Bay Area Air Quality Management District (BAAQMD). The fourth site at a temporary location in the Mountain View Sanitary District. Martinez is specifically located to sample industrial emissions. The Concord site is near the intersection of two major streets

with a combined daily traffic count of approximately 50,000 in a residential and commercial area. The Richmond site is close to a major city street with a daily traffic count of 30,000. Industry is located 3 km miles west of the site. The Pittsburg site is adjacent to a roadway with a daily traffic count of 10,000 and is about 1 km south of an oil burning electrical power plant. The Martinez site is located about 600 m from a petroleum refinery complex which is to the north and west. Approximately 250 m east of the site is a freeway where the daily traffic counts is 60,000. Residential tracts are also nearby.

At the three permanent stations, the samplers were placed on the roof tops of one story buildings, approximately 8-10 m vertically and 25-40 m horizontally from the nearest roadway. At Martinez the samplers were at ground level (1 m). Each location had two hi-vol samplers and one dichotomous sampler to collect particulates for chemical and mutagenic analysis. Gaseous pollutants (CO, SO_2, NO, NO_2 and O_3) were also measured. During the 36 hour episodes, separate 12 hour daytime (0600-1800 and nighttime (1800-0600) samples were collected in order to compare diurnal differences.

Air particulate material for mutagenic and PAH testing was collected on glass fiber filters (Whatman) in standard hi-vol samplers. The filters were used as supplied from the manufacturer and were not pre-treated in any way. Filter-solvent blanks were routinely assayed for mutagenicity and the responses were below detection. Dichotomous fine (<2.5 μmd_a) and coarse (2.5 μm - 15 μmd_a) fraction particulate samples were collected for multielement analysis on 37 mm Teflon Fluoropore (0.2 micron) filters (Ghia) in standard dichotomous samplers (Anderson and Sierra Models).

2. Meteorological Measurements

Temperature and inversion conditions in Contra Costa County during the episodes were inferred from data collected at the Oakland Airport, which is located approximately 25 km from the nearest sampling station. Oakland measurements were made twice daily, at 0400 and 1600 hours PST. In

addition, hourly average wind speeds and wind directions were obtained at Martinez. These meteorological data permitted quantitative characterization of weather conditions but were insufficient to permit accurate descriptions at individual sampling sites. Consequently upwind-downwind relationships to roadways adjacent to the sites could not be established.

3. Chemical Analysis

Air pollutant variables are defined in Table I-1 and the methods used listed in Table I-2. Measurement of trace elements (e.g., Pb, Zn, Fe, Ni) on fine and coarse particulate samples collected with dichotomous aerosol samplers was done by x-ray fluorescence analysis (42). Analyses of the standard particulate pollutants (TSP, $SO_4^=$, NO_3^-, Organics) collected on hi-vol filters were carried out as previously described (18). Gaseous pollutants were continuously monitored using specific gas monitors; O_3 was measured by ultraviolet absorption, CO by infrared absorption, NO and NO_2 by chemiluminescence and SO_2 by fluorescence detection. All methods are EPA reference or equivalent to the EPA reference methods (28,31).

PAH were determined as previously described (18). Sample clean-up steps were omitted, with no loss in resolution. Filters were extracted ultrasonically in trisolvent (toluene:methylene chloride:methanol(1:1:1)) (MCB, Omni-Solv). PAH were separated by HPLC and identified by specific fluorescence and ultraviolet absorption. In addition, the presence of benzanthrone (7-H-benz(de)anthracene-7-one) was confirmed by mass spectral analysis (18).

4. Mutagenicity Testing Methods

Following collection, filters from episode sampling were stored for up to three months at less than $-10^{o}C$ in the dark. Standard methods for extracting air particulate material from filters for mutagenicity testing were used (18). Extractions with trisolvent were carried out under reduced light in an ultrasonic bath and extract residues redissolved in dimethyl sulfoxide (DMSO) for mutagenic analysis. Extracts were stored for 24-48

hours at -10°C. The standard plate incorporation Salmonella/mammalian microsome test was used (12). Mutagenic responses were determined both with and without rat liver homogenate (S9) in strain TA98 which responds mainly to frame-shift mutagens and in TA98NR, a nitroreductase deficient derivative (36). A commercial preparation (Litton Bionetics) of Aroclor 1254 induced rat liver S9 was used. Direct-acting mutagens are detected without S9 and both direct-and indirect-acting mutagens are detected in the presence of S9, although the activities of some direct-acting chemicals are decreased by the addition of S9. The term indirect mutagenicity operationally defines the response of the Ames test in the presence of S9. Ames test results were reported as mutagenic density (revertants produced by the extract from the particles in one m^3 of air) or mutagenic specific activity (revertants per µg benzene soluble organics). Reduced responses of air extracts in TA98NR suggest contributions from nitroarenes.

5. Statistical Methods

Statistical analysis was based on programs contained in the Statistical Analysis System (SAS) (43) run through the California State Health and Welfare Data System.

Correlation analysis was done to relate mutagenicity and PAH variables with selected chemical pollutants. Emphasis was on fine fraction aerosol variables since mutagens are found on small particles (<2.5 μmd_a).

Factor analysis was used to help identify principal types of emission sources. Factor analysis was carried out using the principal component method on a correlation matrix of selected variables (fine fraction trace element concentrations, NO_3^-, mutagenicity and PAH variables). After several preliminary trials, factors with a minimum eigen-value of 0.7 were chosen to be induced in the principal factors. The principal factors retained with this criterion were then used in a varimax rotation procedure.

C. Results and Discussion

1. Meteorological Conditions during Episodes

As noted above, temperature and inversion information were collected twice daily (at 0400 and 1600 hours PST) at the Oakland Airport while wind speed and wind direction were measured at the Martinez sampling site. The wind direction/wind speed data at Martinez are included in Appendix I. San Francisco Bay Area weather factors measured during the episodes by the Bay Area Air Quality Management District are also provided in Appendix II. These data permit the following qualitative descriptions of meteorological conditions prevailing during each episode.

Episode I

Sampling was carried out from 0600 on August 23, to 1800 on August 24, 1982. Two day and one nighttime periods were sampled. At Martinez, winds were from the west throughout the episode at speeds averaging 11 mph by day and 8 mph by night. Oakland surface temperatures were relatively cool reaching a daytime maximum of only 69^{o}F. The minimum was 59^{o}F at night. The base of a shallow inversion at Oakland was 262 m at 0400 hours PST August 23 and 503 m at 1600 hours PST August 24.

Episode II

Two night and one daytime periods were sampled, beginning at 1800 on October 12 and ending at 0600 on October 14, 1982. At Martinez, winds were very light (2-4 mph) throughout and from the south-west during the first night, shifting to the east during the day and becoming westerly during the second night; a daytime surface temperature maximum of 76^{o}F was recorded. The minimum was 52^{o}F. Oakland inversion data were limited; at 0400 hours PST October 13 and 1600 hours PST October 14 the inversion base was at the surface.

Episode III

Two night and one daytime periods were sampled, beginning at 1800 on May 17 and ending at 0600 on May 19, 1983. This episode was carried out during a period of high insolation. Winds were light (3-4 mph) and from the west throughout at Martinez. The Oakland inversion base was 162 m at 0400 hours PST May 17, at the surface at 1600 hours PST May 18 and 66 m at 0400 hours PST May 18. The maximum and minimum surface temperatures at Oakland were 73°F and 55° respectively.

Episode IV

Two night and one daytime periods were sampled, beginning at 1800 on September 12, 1983 and ending at 0600 on September 14, 1983. Westerly breezes prevailed at Martinez throughout the episode, averaging 2 mph during the first night and 6-7 mph during the remaining periods. The base of the Oakland inversion was at the surface at 1600 hours PST, and 0400 hours PST September 12, and again at 1600 hours PST on September 13. Oakland surface temperatures were hot (94°F) just prior to the start of sampling (1500 hours PST, September 12) and fell to 59°F near the end of the period.

Episode V

Two night and one daytime periods were sampled, beginning at 1800 on October 4 and ending at 0600 on October 6, 1983. Again light westerly winds prevailed at Martinez throughout with the Oakland surface temperature reaching a daytime maximum of 76°F and falling to a minimum of 58°F at night. At 0400 hours PST on October 4 the inversion base was 651 m, at 1600 hours PST on October 5 the inversion base was 110 m.

Episode VI

In the final episode, two night and one daytime periods were sampled. Sampling was carried out from 1800 on January 4 to 0600 on January 6, 1984. Martinez winds averaged 5-7 mph and were from the east throughout. Oakland surface temperatures were cool with a maximum of 56°F and a

minimum of 46°F. Oakland inversion data were: 0400 hours PST January 4, base = 181 m; 0400 hours PST January 5, base = surface; 0400 hours PST January 5, base = 89 m.

Episode Summary

Considering the six episodes as a whole, one generality concerning meteorology emerged. With the exception of episode VI, the overall direction of the surface winds was from the west so areawide transport of pollution should be from Richmond, on the west, through Martinez towards Concord and Pittsburg on the east.

2. Combined Episode Data with Diurnal Comparisons

Initially we combined all results of air pollution measurements made during the six intensive sampling episodes in 1982-1984 for statistical analysis. The combined data set contained 72 observations of mutagenicity and chemical pollutant measurements. These data were separated into daytime and nighttime periods for diurnal comparison. Because of the sampling strategy, more observations were made at night (N=44) than during the day (N=28). At the outset, our strategy in sampling episodes was to collect at least one daytime and one nighttime sample. Therefore, we sampled for 36, instead of 24 hours, to improve the chances of obtaining a complete set of samples for two consecutive 12 hour periods. The consequence of having collected samples over 3 consecutive periods was that we analyzed all samples and subsequently have included all sample test results in the statistical analysis. The advantage of using all the results is that we have added one-third more observations to the data base, a substantial increase. The disadvantage is that the data do not contain equal periods of day and night.

Therefore, to calculate means for the combined data based on equal periods of day and night, results of the "twice-sampled" (usually the nighttime period) were averaged and then combined with results of the "once-sampled" period. The method of treating this inequality in this

report is different than the method used in the first report on mutagenicity in Contra Costa County (18). The different methods are as follows:

- Present report: $\text{Mean} = \dfrac{\frac{D + d}{2} + N}{2}$

  where D, d are daytime values and N is a nightime value

- Previous report: $\text{Mean} = \dfrac{D + d + N + N}{4}$.

  where N, the "once-sampled" period is entered twice

Both methods give the same mean values; however the ranges obtained using the present method are reduced somewhat due to the averaging procedure. For example, in Table III-1, the maximum value of 44 rev/m^3 is listed for combined episode data even though during one 12 hour period a value of 58 rev/m^3 was measured.

For correlation and factor analysis the unmodified data were used. Since there are more nighttime than daytime observations, the correlations and factor patterns for the combined episode data reflect larger contributions from nighttime sources.

Summary Statistics

Mean concentrations and other summary statistics for the six episodes combined are shown in Table III-1. Note that the typical sample size shown in the tables (N = 24) is smaller than the actual number of samples collected because of the averaging procedure used to calculate the summary statistics. The 1981-82 (three) episode statistics for the air pollution variables discussed below are shown in Table III-2 so the differences with time can be compared. Variables which are statistically significantly different between the two studies ($p \leq 0.05$), are indicated with an asterisk in Table III-I. (To test the equality of means for mutagens densities and other pollutants between 1981-1982 episodes and 1982-1984

TABLE III-1

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM SIX EPISODES: COMBINED DATA, 1982-1984

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	24	21*	11	3	44
M398MS9	rev/m^3	24	9	5	1	21
ORG98PS9	rev/µg	23	10*	8	<1	30
ORG98MS9	rev/µg	23	5*	4	<1	14
M398NRM	rev/m^3	16	4	1	2	7
NR/98M3	-	16	0.36	0.14	0.15	0.68
BAP	ng/m^3	24	0.2*	0.2	0.1	0.8
BKF	ng/m^3	24	0.1	0.1	0.1	0.4
BGP	ng/m^3	24	0.9	0.6	0.2	2.6
COR	ng/m^3	24	0.6	0.4	0.1	1.5
BZO	ng/m^3	24	0.8*	0.7	0.1	3.2
ORG	µg/m^3	23	3.5*	2.4	1.5	8.4
TSP	µg/m^3	23	64*	21	24	124
NO_3^-	µg/m^3	23	7.9*	4.0	3.2	18.2
$SO_4^=$	µg/m^3	23	8.6*	4.4	5.0	22.3
BRF	ng/m^3	24	45	29	9	117
PBF	ng/m^3	24	242	153	52	605
BRF/PBF	-	24	0.20	0.08	0.13	0.41
ZNF	ng/m^3	24	26*	13	9	68
KF	ng/m^3	24	142	103	50	429
FEF	ng/m^3	24	128	88	26	357
SIF	ng/m^3	24	291	235	56	952
CLF	ng/m^3	24	260	426	27	2173
NIF	ng/m^3	24	7*	6	2	27
SF	ng/m^3	24	1797	1195	516	6473
CO	ppm	18	1.1	0.4	0.5	1.7
NO	pphm	21	1.9	1.2	0.3	4.3
NO_2	pphm	23	2.6*	1.1	0.9	4.9
O_3	pphm	23	2.2	1.1	0.1	4.1
SO_2	pphm	23	0.4	0.7	0.0	3.4

*Mean significantly different ($p \leq 0.05$) from mean during 1981-82 episodes.

TABLE III-2

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM THREE EPISODES: COMBINED DATA 1981-1982

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	12	12	6	6	21
M398MS9	rev/m^3	12	7	2	3	10
ORG98PS9	rev/mg	12	2	3	<1	8
ORG98MS9	rev/mg	12	1	1	<1	3
M398NRM	rev/m^3	12	3	1	<1	5
NR/98M3	-	12	0.43	0.16	0.18	0.71
BAP	ng/m^3	12	0.6	0.6	0.1	1.6
BKF	ng/m^3	12	0.3	0.2	0.1	0.7
BGP	ng/m^3	12	1.4	1.1	0.4	3.4
COR	ng/m^3	12	0.8	0.5	0.2	1.9
BZO	ng/m^3	12	2.1	2.0	0.3	5.8
ORG	ng/m^3	12	7.5	3.5	2.0	10.7
TSP	$\mu g/m^3$	12	90	22	52	126
NO_3	$\mu g/m^3$	12	11.5	4.6	4.1	18.5
SO_4	$\mu g/m^3$	12	14.9	5.7	5.3	25.2
BRF	ng/m^3	11	69	48	16	172
PBF	ng/m^3	11	262	162	82	671
BRF/PBF	-	11	0.25	0.07	0.15	0.40
ZNF	ng/m^3	11	37	13	12	55
KF	ng/m^3	NA	-	-	-	-
FEF	ng/m^3	11	102	39	42	155
SIF	ng/m^3	NA	-	-	-	-
CLF	ng/m^3	NA	-	-	-	-
NIF	ng/m^3	11	25	14	7	51
SF	ng/m^3	NA	-	-	-	-
CO	ppm	9	1.2	0.5	0.6	1.9
NO	pphm	9	2.8	2.2	0.7	6.3
NO_2	pphm	9	3.6	1.5	1.0	6.1
O_3	pphm	9	1.6	1.4	0.1	4.0
SO_2	pphm	9	0.4	0.3	0	0.9

*NA = Not analyzed.

episodes, t-tests were carried out. Appropriate t-statistics were chosen based on the results of F-tests on equal variances. If the variances were equal, t-statistics derived from pooled variances were used. Otherwise, t-statistics derived from separate variances were used.)

The combined six episode mean for mutagenic density in TA98 was 21 rev/m^3 (with S9) and 9 rev/m^3 (without S9). Thus both direct and indirect acting mutagens are present. The value with S9 is significantly higher than the mutagenic density previously measured during pollution episodes in 1981-82 (12 rev/m^3 +S9) (18). In the present study, the mean mutagenic density in the nitroreductase deficient strain, TA98NR (-S9) was 4 rev/m^3 and the ratio of TA98NR/TA98 was 0.36; both values are similar to those measured in the earlier study. Thus more than half of the mutagenic activity of aerosol extracts is dependent upon enzymatic nitroreduction. This implies that mononitroorganics, such as 1-nitropyrene which are known to be present in community aerosols elsewhere (15,17), probably make major contributions to the mutagenicity of Contra Costa aerosols.

Mean mutagenic specific activities (rev/µg ORG) were 10 (+S9) and 5 (-S9). These are both significantly higher values than those measured in 1981-82. (cf Table III-2) Note that significantly lower benzene soluble organic concentrations were also found in the present study. The mean organic concentration measured (3.5 µg/m^3) was approximately a factor of two lower than that measured in the 1981-82 episodes (7.5 µg/m^3). Thus, although the organics in general have dropped, the organics that remain are much more mutagenic. Among PAH, levels of BAP and BZO were also significantly lower than previously measured. In the present study, concentrations ranged from the detection limit (0.1 ng/m^3) for BKF and 0.2 ng/m^3 for BAP to 0.9 ng/m^3 for BGP.

The mean episode TSP level in the present study was 64 µg/m^3, significantly lower than previously found. In 1981-82, the episode mean TSP value was 90 µg/m^3. These results indicate that mutagenic density has increased, despite decreasing TSP and aerosol organic levels. Increasing mutagenic specific activity over time is of potential concern to public health and is analyzed in greater detail in Chapter IV.

Mean concentrations of NO_3^- and $SO_4^=$ were 7.9 and 8.6 $\mu g/m^3$ respectively, also significantly lower (by approximately 40 percent) than those observed in 1981-82. The Hi-Vol $SO_4^=$ concentration was comparable to the $SO_4^=$ value calculated from the fine fraction sulfur concentration (1.8 $\mu g/m^3$). (Only about 10 percent of S (0.2 $\mu g/m^3$) was found in the coarse fraction.) Assuming all of the fine S is in the form of $SO_4^=$, the mean fine fraction $SO_4^=$ concentration was calculated to be approximately 5.4 $\mu g/m^3$, or two-thirds the amount of $SO_4^=$ found by the Hi-vol method.

Among gaseous pollutants, the mean CO concentrations was 1.1 ppm. Means of NO, NO_2 and O_3 were, 1.9, 2.6 and 2.2 pphm respectively. The mean SO_2 concentration was 0.4 pphm. These gas concentrations are similar to those measured earlier in Contra Costa, although NO_2 concentrations were significantly lower. Pitts and coworkers have recently described a possible filter sampling artifact related to O_3 (23). Increased mutagenicity was measured when aerosols were collected on glass fiber filters in the presence of higher O_3 concentrations (> 10 pphm). However, O_3 concentrations measured in Contra Costa County were all below those which produced significant artifacts in the study of Pitts et al, which was carried out in El Monte and Riverside.

Among aerosol trace elements, fine fraction lead concentration was 242 ng/m^3, very near to the mean concentration measured in 1981-82 episodes (262 ng/m^3). Fine fraction Br was 45 ng/m^3 and the Br/Pb ratio was 0.2, indicating the presence of an aged aerosol. Ratios in fresh auto emissions are typically greater than 0.3. Fine fraction Zn was 26 ng/m^3, significantly below the 1981-82 value (37 ng/m^3). The fine fraction iron concentration (128 ng/m^3) was comparable to the 1981-82 value (102 ng/m^3). The fine fraction Ni concentration was 25 ng/m^3 in the previous study and 7 ng/m^3 in the present investigation. We can provide no explanation for the significant threefold decrease in Ni. Among other trace elements, the mean fine fraction potassium concentration was 142 ng/m^3. The K/Fe ratio of 1.1 is higher than typically seen in soil (0.5), but much lower than in aerosols derived primarily from wood combustion ($\geq$8) (44).

For most variables, the diurnal differences (cf Tables III-3 and 4) were small. Mutagenic density (+S9) was slightly higher by day (24 rev/m^3) than by night (17 rev/m^3). However, direct-acting (-S9) mutagenic density was nearly constant from day (10 rev/m^3) to night (9 rev/m^3). Organic levels (total and specific PAH) were also very similar from day to night. TSP and NO_3^- were both slightly higher by day while $SO_4^=$ showed essentially no diurnal change.

Two measured pollutants, CLF and O_3, exhibited clear diurnal differences. Fine fraction chloride (CLF) was twice as high at night while O_3 was twice as high by day. (cf Tables III-3,4). The difference in CLF may be related to diurnal differences in relative humidity. The O_3 difference reflected daytime photochemical formation of ozone in the atmosphere.

Correlation Analysis

Correlation analysis was carried out to explore relationships between mutagens, PAH and source emissions tracers. Correlations between mutagenic density, PAH and selected elements and gases are shown in Tables III-5-7. (Complete correlation matrices are provided in the Appendix III).

Mutagenic density variables (±S9) were very strongly correlated ($p \leq 0.01$) with each other and with PAH. Mutagenicity variables and PAH were also significantly ($p \leq 0.05$) correlated with automotive tracers, BRF and PBF. For the combined episode, as well as day and night data, correlations with BRF were higher than with PBF. Mutagenic density and PAH were also positively correlated with particulate NO_3^- and gaseous CO, NO, NO_2. There were significant negative correlations of mutagenic density with CLF and O_3. PAH were also negatively correlated with O_3.

Among the PAH variables, COR was very highly correlated ($p \leq 0.01$) with CO, PBF, and BRF, all three considered primarily automotive pollutants. COR was also correlated with NO and NO_2 and KF. In other studies, KF has been identified as a wood smoke tracer (44). Although not shown in the tables, correlations of BKF were like BAP and BGP like COR.

TABLE III-3

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM SIX EPISODES: DAYTIME SAMPLES, 1982-1984

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	24	24	14	6	58
M398MS9	rev/m^3	24	10	6	2	27
ORG98PS9	rev/µg	23	12	10	<1	39
ORG98MS9	rev/µg	23	5	5	<1	8
M398NRM	rev/m^3	16	4	2	1	7
NR/98M3	-	16	0.35	0.22	0.12	1.10
BAP	ng/m^3	24	0.2	0.1	0.1	0.4
BKF	ng/m^3	24	0.1	0.1	0.1	0.3
BGP	ng/m^3	24	1.1	0.7	0.2	2.4
COR	ng/m^3	24	0.7	0.4	0.1	1.6
BZO	ng/m^3	24	0.8	0.6	0.1	2.0
ORG	$\mu g/m^3$	23	3.6	2.7	1.5	9.2
TSP	$\mu g/m^3$	23	76	25	34	139
NO_3^-	$\mu g/m^3$	23	8.6	3.4	3.2	15.8
$SO_4^=$	$\mu g/m^3$	23	8.4	4.4	5.1	21.4
BRF	$\mu g/m^3$	24	50	31	8	114
PBF	$\mu g/m^3$	24	266	157	49	638
BRF/PBF	-	24	0.19	0.07	0.10	0.42
ZNF	ng/m^3	24	31	24	9	123
KF	ng/m^3	24	157	129	38	491
FEF	ng/m^3	24	144	98	28	371
SIF	ng/m^3	24	326	284	24	1220
CLF	ng/m^3	24	174	282	19	1450
NIF	ng/m^3	24	8	8	2	33
SF	ng/m^3	24	1810	1384	442	7640
CO	ppm	18	1.3	0.4	0.6	2.1
NO	pphm	21	2.2	1.5	0.5	6.4
NO_2	pphm	23	2.6	1.1	1.2	5.3
O_3	pphm	23	2.9	1.5	0.0	5.9
SO_2	pphm	23	0.4	0.9	0.0	4.3

TABLE III- 4

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM
SIX EPISODES:
NIGHTTIME SAMPLES, 1982-1984

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	24	17	10	<1	37
M398MS9	rev/m^3	24	9	5	<1	18
ORG98PS9	rev/µg	24	9	6	<1	21
ORG98MS9	rev/µg	24	4	3	<1	10
M398NRM	rev/m^3	16	4	2	1	6
NR/98M3	-	16	0.36	0.13	0.18	0.70
BAP	ng/m^3	24	0.2	0.3	0.1	1.5
BKF	ng/m^3	24	0.1	0.1	0.1	0.5
BGP	ng/m^3	24	0.8	0.7	0.2	2.8
COR	ng/m^3	24	0.5	0.3	0.1	1.5
BZO	ng/m^3	24	0.7	1.0	0.1	4.4
ORG	$\mu g/m^3$	24	3.3	2.3	1.1	8.8
TSP	$\mu g/m^3$	24	52	21	15	109
NO_3^-	$\mu g/m^3$	24	7.1	4.9	3.2	20.5
$SO_4^=$	$\mu g/m^3$	24	8.6	4.4	5.0	23.1
BRF	ng/m^3	24	41	30	7	133
PBF	ng/m^3	24	218	168	38	714
BRF/PBF	-	24	0.21	0.10	0.11	0.41
ZNF	ng/m^3	24	20	11	3	52
KF	ng/m^3	24	127	82	54	377
FEF	ng/m^3	24	112	90	24	353
SIF	ng/m^3	24	256	205	87	787
CLF	ng/m^3	24	346	582	31	2895
NIF	ng/m^3	24	7	7	2	30
SF	ng/m^3	24	1785	1094	590	5305
CO	ppm	18	1.0	0.4	0.2	1.7
NO	pphm	21	1.5	1.3	0.1	5.0
NO_2	pphm	23	2.5	1.2	0.6	5.1
O_3	pphm	23	1.5	0.9	0.1	3.5
SO_2	pphm	23	0.3	0.6	0.0	2.5

TABLE III- 5

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS: COMBINED EPISODE DATA, 1982-1984

	TA98+S9	TA98-S9	BAP	COR	BZO
TA98+S9	1.00	0.94**	0.45**	0.54**	0.60**
TA98-S9	0.94**	1.00	0.49**	0.44**	0.59**
BAP	0.45**	0.49**	1.00	0.38**	0.92**
COR	0.54**	0.44**	0.37**	1.00	0.56**
BZO	0.60**	0.59**	0.92**	0.56**	1.00
PBF	0.28*	0.25*	0.08	0.71**	0.19
BRF	0.40**	0.38**	0.30**	0.74**	0.44**
KF	0.26*	0.22	0.15	0.58**	0.08
ZNF	0.12	0.12	-0.01	0.27*	0.26
FEF	0.09	0.13	-0.07	0.28*	-0.05
SIF	-0.12	0.01	-0.07	0.13	-0.09
CLF	-0.27*	-0.22	0.01	-0.15	-0.05
NIF	-0.10	-0.07	-0.12	-0.20	-0.12
SF	0.22	0.23*	0.28*	0.03	0.36**
NO_3^-	0.49**	0.51**	0.32**	0.15	0.47**
CO	0.54**	0.51**	0.23	0.71**	0.36**
NO	0.51**	0.45**	0.34**	0.49**	0.44**
NO_2	0.32**	0.31**	0.11	0.61**	0.18
O_3	-0.27*	-0.35**	-0.41**	-0.13	-0.43**
SO_2	0.04	0.05	-0.02	-0.27*	-0.02

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

TABLE III-6

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS: DAYTIME SAMPLES, 1982-1984

	TA98+S9	TA98-S9	BAP	COR	BZO
TA98+S9	1.00	0.95**	0.51**	0.50**	0.65**
TA98-S9	0.95**	1.00	0.57**	0.38*	0.59**
BAP	0.53**	0.57**	1.00	0.51**	0.77**
COR	0.50**	0.38*	0.51**	1.00	0.76**
BZO	0.67**	0.59**	0.77**	0.76**	1.00
PBF	0.19	0.07	0.28	0.70**	0.46*
BRF	0.38*	0.30	0.59**	0.74**	0.71**
KF	0.24	0.10	0.39*	0.66**	0.48*
ZNF	-0.14	-0.19	-0.19	0.15	-0.01
FEF	-0.12	-0.18	0.04	0.19	0.03
SIF	-0.12	-0.16	0.14	0.15	0.05
CLF	-0.20	-0.12	-0.08	-0.12	-0.06
NIF	-0.33	-0.39*	-0.19	-0.33	-0.24
SF	0.23	0.28	0.54**	0.20	0.45*
NO_3^-	0.56**	0.57**	0.66**	0.45*	0.64**
CO	0.53**	0.42	0.32	0.65**	0.54*
NO	0.49*	0.49*	0.46*	0.12	0.45*
NO_2	0.24	0.12	0.19	0.51**	0.26
O_3	-0.30	-0.36	-0.63**	-0.03	-0.48*
SO_2	-0.03	0.01	0.23	-0.33	0.00

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

TABLE III- 7

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS:
NIGHTTIME SAMPLES, 1982-1984

	TA98+S9	TA98-S9	BAP	COR	BZO
TA98+S9	1.00	0.95**	0.56**	0.55**	0.66**
TA98-S9	0.95**	1.00	0.56**	0.50**	0.63**
BAP	0.56**	0.56**	1.00	0.46**	0.96**
COR	0.55**	0.50**	0.46**	1.00	0.54**
BZO	0.66**	0.63**	0.96	0.54**	1.00
PBF	0.33*	0.35*	0.06	0.73**	0.13
BRF	0.43**	0.43**	0.28	0.77**	0.37*
KF	0.27	0.33*	0.14	0.53**	0.18
ZNF	0.34*	0.47**	0.10	0.28	0.15
FEF	0.21	0.32*	-0.09	0.33*	-0.08
SIF	0.05	0.16	-0.12	0.09	-0.15
CLF	-0.31*	-0.28	0.00	-0.15	-0.05
NIF	0.10	0.19	-0.12	-0.13	-0.07
SF	0.22	0.20	0.26	-0.08	0.35*
NO_3^-	0.49**	0.51**	0.30*	0.00	0.44**
CO	0.55**	0.58**	0.28	0.72**	0.34
NO	0.51**	0.42**	0.38*	0.71**	0.47**
NO_2	0.40**	0.43**	0.10	0.70**	0.16
O_3	-0.50**	-0.47**	-0.41**	-0.50**	-0.53**
SO_2	0.01	0.09	-0.09	-0.27	-0.04

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

Factor Analysis

Further attempts to characterize major sources of mutagenic air pollution were carried out using principal component factor analysis. Because of the large number of pollution variables measured, interpretation of sources is difficult. The advantage of factor analysis is that it groups the variables into a smaller number of factors which explain a large portion of the overall variance in the data set. Variables in each factor may have a common source or formation process (23). Ten aerosol variables were used, including tracers for a variety of sources. They were PBF and BRF (automotive), FEF and SIF (soil), NIF (industry; oil refining), CLF (marine influence), NO_3^- (secondary aerosols), SF (industry; secondary $SO_4^=$), ZNF (many possible sources; traffic, industry, incineration) and KF (possible agricultural burning or wood smoke indicator). Gas pollutant and meteorological results were excluded because of limited or missing data. Factor analysis was not tried using the available meteorological and gas pollutant data.

Approach I: Factor Patterns of Major Source Emissions Tracers, Excluding Mutagenic Density and PAH Variables.

Consider first the factor patterns obtained without mutagenic density or PAH variables (Tables III-8-10). Using the ten aerosol source tracers in the combined episode data set, four factors were present, which accounted for 81 percent of the total variance (cf Table III-8). The most important factor (No. 1) was heavily loaded with automotive (PBF, BRF) and soil (FEF, SIF) tracers and explained 43 percent of the variance. KF was also strongly associated with Factor No. 1. The second factor contained tracers for industry (NIF) as well as soil and explained 16 percent of the variance. The third factor contained tracers for secondary aerosols (NO_3^- and SF) and explained 12 percent of the variance and the fourth factor contained the marine source tracer (CLF), and explained 10 percent of the variance. ZNF was distributed over all four factors implying that zinc was derived from many sources.

TABLE III-8

PRINCIPAL COMPONENT FACTORS FOR PARTICULATE AIR POLLUTANTS: COMBINED EPISODE DATA, 1982-1984 (N = 71)

	Factor 1	Factor 2	Factor 3	Factor 4	
	PBF (0.95)*	NIF (0.88)	SF (0.84)	CLF (0.96)	
	BRF (0.93)	SIF (0.63)	NO_3^- (0.83)	ZNF (0.30)	
	KF (0.90)	FEF (0.61)	ZNF (0.25)		
	FEF (0.71)	ZNF (0.42)			
	SIF (0.53)				
	ZNF (0.48)				
Variance Explained (%)	43	16	12	10	Sum: 81
Interpretation:	Automotive Emissions and Soil	Oil Refining and Soil	Secondary Aerosols	Sea Salt	

Zinc distributed in all four factors.

*Factor loadings $\geq$0.25 are shown in parenthesis.

TABLE III-9

PRINCIPAL COMPONENT FACTORS FOR PARTICULATE AIR POLLUTANTS: DAYTIME SAMPLES, 1982-1984 (N = 27)

	Factor 1	Factor 2	Factor 3	Factor 4	
	KF (0.92)*	CLF (0.94)	NIF (0.87)	SF (0.96)	
	PBF (0.90)	ZNF (0.90)	SIF (0.38)	BRF (0.38)	
	FEF (0.83)	NO_3^- (0.36)	FEF (0.37)	NO_3^- (0.31)	
	BRF (0.81)				
	SIF (0.80)				
	NO_3^- (0.28)				
Variance Explained (%)	40	18	17	10	Sum: 85
Interpretation:	Automotive Emissions and Soil	Sea Salt and Zinc	Oil Refining and (some) Soil	Secondary $SO_4^=$ Aerosols	

Nitrate found in three factors

*Factor loadings $\geq$0.25 are shown in parenthesis.

TABLE III-10

PRINCIPAL COMPONENT FACTORS FOR PARTICULATE AIR POLLUTANTS: NIGHTTIME SAMPLES, 1982-1984 (N = 44)

	Factor 1	Factor 2	Factor 3	Factor 4	
	BRF (0.95)*	NIF (0.81)	SF (0.87)	CLF (0.99)	
	PBF (0.93)	SIF (0.80)	NO_3^- (0.87)		
	KF (0.89)	FEF (0.66)	NIF (0.34)		
	ZNF (0.72)	ZNF (0.34)	ZNF (0.28)		
	FEF (0.67)	KF (0.27)			
	SIF (0.40)				
Variance Explained (%)	48	18	11	9	Sum: 86
Interpretation:	Automotive Emissions, Zinc and Soil	Oil Refining and Soil	Secondary Aerosols	Sea Salt	

Zinc found in three factors

*Factor loadings $\geq$0.25 are shown in parenthesis.

Both the day and nighttime factor patterns contain the same four factors, with slight modifications (cf Tables III-9,10). By day, the automotive-soil factor (No. 1) explained 40 percent of the variance; by night this factor explained 48 percent of the variance and was also heavily loaded with ZNF. During the day, the sea salt (CLF) factor (No. 2) contained some ZNF and explained 18 percent of the variance. At night, CLF was isolated in a factor (No. 4), which contained only 9 percent of the variance. Also, during the day NO_3^--associated aerosols are spread over three factors (No. 1 "Auto-soil", No. 2 "Sea salt-zinc", No. 4, "Fine sulphur secondary"); at night, NO_3^- (secondary aerosol) is found together with fine sulphur (SF) in factor No. 3. This nighttime secondary aerosol factor explained 11 percent of the total variance.

Approach II: Factor Patterns of Major Source Emissions Tracers, Including Mutagenic Density and PAH Variables

Factor analysis was repeated including one additional mutagenic density or PAH variable at a time. The factor patterns with mutagenic density, coronene and benzanthrone were as follows:

i. Mutagenic Density

With the addition of mutagenic density (in TA98 +S9) to the combined episode data base, five factors were obtained instead of four (compare Tables III-8 and III-11). Factors loaded with tracers for auto emissions and soil (No. 1), oil refining and soil (No. 2) and sea salt (No. 4) were again obtained. However with the mutagenic density variable entered, NO_3^- was heavily loaded in one factor (No. 3), while SF was heavily loaded in another (No. 5). Mutagenicity was most strongly associated with the NO_3^- secondary aerosol factor (No. 3) (loading = 0.68) and also with the auto-soil factor (No. 1) (loading = 0.29).

TABLE III-11

PRINCIPAL COMPONENT FACTORS FOR PARTICULATE AIR POLLUTANTS: COMBINED EPISODE DATA INCLUDING MUTAGENIC DENSITY (+S9)

	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	
	PBF (0.94)*	NIF (0.84)	NO_3^- (0.82)	CLF (0.94)	SF (0.90)	
	BRF (0.93)	FEF (0.60)	ZNF (0.52)	ZNF (0.30)	NO_3^- (0.35)	
	KF (0.90)	SIF (0.60)	SF (0.26)		NIF (0.27)	
	FEF (0.72)	ZNF (0.50)				
	SIF (0.56)					
	ZNF (0.37)					
Mutagenic Density (+S9)	(0.29)	(-0.29)	(0.68)	(-0.41)	(0.08)	
Variance Explained (%)	39	18	11	10	6	Sum: 85
Interpretation:	Automotive Emissions and Soil	Oil Refining and Soil	Secondary (NO_3^-) and Zinc	Sea Salt	Secondary ($SO_4^=$)	

*Factor loadings $\geq$0.25 are shown in parenthesis.

TABLE III-12

PRINCIPAL COMPONENT FACTORS FOR PARTICULATE AIR POLLUTANTS: DAYTIME SAMPLES INCLUDING MUTAGENIC DENSITY (+S9)

	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	
	PBF (0.95)*	CLF (0.95)	NO_3^- (0.87)	NIF (0.86)	SF (0.96)	
	KF (0.92)	ZNF (0.91)	BRF (0.31)	SIF (0.53)	BRF (0.30)	
	BRF (0.84)	NO_3^- (0.26)		FEF (0.52		
	FEF (0.75)					
	SIF (0.71)					
Mutagenic Density (+S9)	(0.07)	(-0.24)	(0.83)	(-0.24)	(0.13)	
Variance Explained (%)	36	21	16	9	7	Sum: 89
Interpretation:	Automotive Emissions and Soil	Sea Salt and Zinc	Secondary (NO_3^-)	Oil Refining and Soil	Secondary ($SO_4^=$)	

*Factor loadings $\geq$0.25 are shown in the parenthesis.

TABLE III- 13

PRINCIPAL COMPONENT FACTORS FOR PARTICULATE AIR POLLUTANTS: NIGHTTIME SAMPLES INCLUDING MUTAGENIC DENSITY (+S9)

	Factor 1	Factor 2	Factor 3	Factor 4	
	BRF (0.94)*	NO_3^- (0.88)	NIF (0.81)	CLF (0.91)	
	PBF (0.92)	SF (0.85)	SIF (0.72)		
	KF (0.89)	NIF (0.32)	FEF (0.60)		
	ZNF (0.73)	ZNF (0.28)	ZNF (0.31)		
	FEF (0.71)		KF (0.25)		
	SIF (0.47)				
Mutagenic Density (+S9)	(0.44)	(0.43)	(-0.26)	(-0.59)	
Variance Explained (%)	45	18	11	10	Sum: 84
Interpretation:	Auto Emissions, Zinc and Soil	Secondary	Oil Refining and Soil	Sea Salt	

*Factor loadings ≥ 0.25 are shown in parenthesis.

Day versus Night comparisons were as follows: When mutagenic density was added to the daytime data set, five factors were obtained (cf Table III-12). However, with mutagenicity results in the nighttime data set, only four factors were obtained (cf Table III-13). In the daytime pattern, NO_3^- was associated with one factor (No.3) while SF was associated with another (No. 5). In the nighttime pattern, both NO_3^- and SF were heavily loaded together in the same factor (No. 2).

The association of mutagenic density with the day and night factor patterns provides a striking diurnal comparison which invites interpretation. During the day, mutagenic density was heavily loaded in the NO_3^- factor (No. 3) (loading = 0.83), and not loaded in the auto-soil factor, (No. 1) (loading = 0.07). (cf Table III-12). However during the night mutagenic density was loaded equally into the auto-soil factor (No. 1) (loading = 0.44) and the NO_3^- factor (No. 2) (loading = 0.43). We can speculate on the possible significance of these statistical diurnal differences. The strong loading of mutagenicity in the NO_3^--factor during the day suggests that a higher percentage of mutagenic aerosols may be associated with areawide secondary aerosols.

The association of mutagenicity with NO_3^--containing aerosols especially by day could be the result of aging of primary aerosols containing mutagens to secondary particulate matter. Or it may be related to nitric acid artifacts known to be present in NO_3^- measurements made on glass fiber filters. As described by Appel and co-workers (27), a portion of the NO_3^- measured on glass fiber filters is due to gaseous HNO_3, which binds to reactive (alkaline) sites on the sampling medium. HNO_3 may participate in the formation of highly mutagenic nitroarenes either in the air or on the filters. Furthermore, HNO_3 exhibits a diurnal pattern with maximum values occurring during the day as with O_3 (23). The diurnal behavior of HNO_3 may be related to the present finding

of a strong daytime association between mutagenic density and NO_3^-. Additional research is required to elucidate these complex issues. For example, direct measurement of gaseous HNO_3, as well as aerosol collection with and without a HNO_3 denuder are needed.

In related research, Fitz et al (45) compared POM collected on Hi-Vol samplers equipped with and without a HNO_3 denuder and found that mutagenicity did not depend on the reexposure of aerosols to HNO_3 during collection in Los Angeles. Concentrations of HNO_3 in Contra Costa County are much lower than in Los Angeles, suggesting that gas phase HNO_3 artifacts probably did not significantly influence results of mutagenicity measurements made in the present study.

Finally, negative associations were obtained between mutagenic density and both the "sea salt" and "oil-soil" factors. Negative mutagenicity loadings on these factors were found consistently, in the combined episode, and day and night data (Tables III-11-13). The negative association with CLF may reflect directional transport of pollutants by sea breezes. However the explanation for the negative association of mutagenicity with the "oil-soil" factor is not clear.

ii. Coronene

When the results for coronene were combined with those of ten aerosol tracer pollutants, factor analysis of the combined episode data base gave the pattern shown in Table III-14. (Similar results, not shown, were obtained using either the day or the night data.) In the case of the combined episode data, four factors were resolved which explained 81 percent of the variance. Automotive emissions (PBF, BRF) were found primarily in factor No. 1, oil (NIF) and soil (FEF, SIF) tracers were in factor No. 2, secondary aerosols (NO_3^-, SF) were in a third and the marine tracer (CLF) in a fourth factor.

TABLE III- 14

PRINCIPAL COMPONENT FACTORS FOR PARTICULATE AIR POLLUTANTS: COMBINED EPISODE DATA INCLUDING CORONENE

	Factor 1	Factor 2	Factor 3	Factor 4	
	BRF (0.91)*	NIF (0.81)	SF (0.84)	CLF (0.97)	
	PBF (0.90)	FEF (0.78)	NO_3^- (0.83)	ZNF (0.30)	
	KF (0.82)	SIF (0.77)	ZNF (0.26)		
	ZNF (0.41)	ZNF (0.46)			
	SIF (0.35)	KF (0.37)			
		PBF (0.32)			
Coronene	(0.89)	(-0.24)	(0.05)	(-0.12)	
Variance Explained (%)	42	15	14	10	Sum: 81
Interpretation:	Automotive Emissions	Oil Refining and Soil	Secondary Aerosols	Sea Salt	

*Factor loadings $\geq$0.25 are shown in parenthesis.

TABLE III-15

PRINCIPAL COMPONENT FACTORS FOR PARTICULATE AIR POLLUTANTS: COMBINED EPISODE DATA INCLUDING BENZANTHRONE

	Factor 1	Factor 2	Factor 3	Factor 4	
	PBF (0.95)*	NO_3^- (0.82)	NIF (0.86)	CLF (0.96)	
	BRF (0.91)	SF (0.80)	SIF (0.52)	ZNF (0.32)	
	KF (0.90)	BRF (0.27)	FEF (0.51)		
	FEF (0.79)		ZNF (0.42)		
	SIF (0.62)				
	ZNF (0.49)				
Benzanthrone	(0.28)	(0.69)	(-0.41)	(-0.10)	
Variance Explained (%)	39	19	11	10	Sum: 79
Interpretation:	Auto Emissions and Soil	Secondary Aerosols	Oil Refining and Soil	Sea Salt	

*Factor loadings $\geq$0.25 are shown in parenthesis.

Coronene was heavily loaded only in factor No. 1, the automotive factor, which explained 42 percent of the variance. Unlike mutagenic density, COR was not associated with the secondary aerosol factor (No. 3). As with mutagenic density, COR was negatively associated with factors containing tracers for oil refining and soil (No. 2) and sea salt (No. 4).

The results of factor analysis using COR amplify the two-variable correlations described above (cf Table III-5-7). COR was found to be strongly associated with elemental tracers for automotive emissions (PBF, BRF), in the combined episode, as well as the day and night data sets. Thus COR was derived primarily from mobile sources during these sampling episodes in Contra Costa county.

iii. Benzanthrone

The factor pattern including benzanthrone with the ten tracer variables is shown in Table III-15. Only the results for the combined episode data base are presented. However, the factor patterns for day and night were similar. Again four factors were resolved, containing tracers for autos and soil (No. 1), secondary aerosols (No. 2), oil refining and soil (No. 3) and sea salt (No. 4). Benzanthrone was associated most strongly with the secondary aerosol factor (loading = 0.69) and, less strongly, with the automotive-soil factor (loading = 0.28). BZO was also negatively associated with the "oil-soil" and "sea salt" factors. Sea breezes presumably accounted for the negative association with CLF.

The observation that, benzanthrone, a carbonyl-arene, was strongly associated with the secondary (NO_3^-) factor, could suggest that BZO is formed by atmospheric oxidation of precursor PAH. Further oxidation of benzanthrone may lead to its destruction and lower concentrations with increasing time and at distances downwind from a source. The association of BZO with the auto emissions factor (No. 1 in Table III-15) suggests that this oxidized PAH is also present in tailpipe emissions.

iv. Summary

In summary, factor analysis revealed that:

1. Mutagenic density was associated with secondary aerosols (NO_3^-) automotive emissions (PBF, BRF) and fine surface soil (FEF, SIF).

2. Coronene was associated with automotive emissions and fine surface soil.

3. Benzanthrone was associated with secondary aerosols, as well as automotive emissions and fine surface soil.

4. Comparing mutagenicity and PAH variables, mutagenic density exhibited a factor pattern more like benzanthrone and unlike coronene. In this respect some mutagens and BZO both behaved like secondary aerosols.

3. Geographic Differences

Summary Statistics

Summary statistics for air pollutants measured at each of the four stations are shown in Tables III-16-19. Mutagenic density and specific activity were highest at Concord (29 rev/m^3, 14 rev/µg respectively), followed by Pittsburg. The lowest values, observed at Martinez, were, 14 rev/m^3 and 7 rev/µg. These mean values were higher at all four stations in comparison with mean values measured during the 1981-82 episode study.

The mutagenic density (-S9) ratio, TA98NR/TA98, was highest at Richmond (0.45) and at least twenty-five percent lower at the other three stations (0.31-0.34). These ratios suggest that NO_2-organics contributed a major fraction of the mutagenic activity observed in the present study. These

TABLE III-16

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM RICHMOND DURING SIX EPISODES 1982-1984

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	18	17	11	4	34
M398MS9	rev/m^3	18	7	5	3	16
ORG98PS9	rev/µg	17	8	8	<1	21
ORG98MS9	rev/µg	17	4	4	<1	10
M398NRM	rev/m^3	12	4	1	3	5
NR/98M3	-	12	0.45	0.17	0.33	0.69
BAP	ng/m^3	18	0.2	0.2	0.1	0.5
BKF	ng/m^3	18	0.1	0.1	0.1	0.3
BGP	ng/m^3	18	1.0	0.6	0.4	1.8
COR	ng/m^3	18	0.7	0.3	0.3	1.0
BZO	ng/m^3	18	0.8	0.7	0.2	2.1
ORG	$\mu g/m^3$	17	3.5	2.5	1.6	7.3
TSP	$\mu g/m^3$	17	57	19	24	70
NO_3^-	$\mu g/m^3$	17	7.8	3.5	4.0	11.8
$SO_4^=$	$\mu g/m^3$	17	10.8	6.6	5.7	22.2
BRF	ng/m^3	18	42	24	9	66
PBF	ng/m^3	18	209	125	52	407
BRF/PBF	-	18	0.21	0.09	0.13	0.37
ZNF	ng/m^3	18	28	23	9	68
KF	ng/m^3	18	125	75	50	247
FEF	ng/m^3	18	88	66	26	188
SIF	ng/m^3	18	155	121	56	393
CLF	ng/m^3	18	497	825	44	2172
NIF	ng/m^3	18	7	5	2	15
SF	ng/m^3	18	2506	2032	1074	6473
CO	ppm	18	1.2	0.2	0.9	1.4
NO	pphm	18	1.5	1.0	0.3	3.0
NO_2	pphm	18	2.3	1.1	1.0	3.9
O_3	pphm	18	2.3	1.1	0.4	3.6
SO_2	pphm	18	0.1	0.2	0.0	0.4

TABLE III-17

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM MARTINEZ DURING SIX EPISODES 1982-1984

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	18	14	8	3	25
M398MS9	rev/m^3	18	7	4	1	12
ORG98PS9	rev/µg	18	7	6	<1	16
ORG98MS9	rev/µg	18	4	3	<1	7
M398NRM	rev/m^3	12	3	1	2	4
NR/98M3	-	12	0.32	0.10	0.18	0.43
BAP	ng/m^3	18	0.2	0.1	0.1	0.2
BKF	ng/m^3	18	0.1	0	0.1	0.1
BGP	ng/m^3	18	0.6	0.3	0.2	1.0
COR	ng/m^3	18	0.3	0.2	0.1	0.7
BZO	ng/m^3	18	0.5	0.3	0.2	0.9
ORG	µg/m^3	18	3.3	2.5	1.6	7.0
TSP	µg/m^3	18	77	29	35	124
NO_3^-	µg/m^3	18	7.9	5.1	3.2	17.8
$SO_4^=$	µg/m^3	18	10.7	5.2	6.8	20.4
BRF	ng/m^3	18	45	28	12	101
PBF	ng/m^3	18	229	193	85	599
BRF/PBF	-	18	0.23	0.11	0.16	0.41
ZNF	ng/m^3	18	24	8	16	40
KF	ng/m^3	18	143	123	64	386
FEF	ng/m^3	18	177	121	38	357
SIF	ng/m^3	18	463	354	120	952
CLF	ng/m^3	18	235	133	61	449
NIF	ng/m^3	18	15	7	6	27
SF	ng/m^3	18	1883	741	846	2915
CO	ppm	-	-	-	-	-
NO	pphm	9	3.1	1.1	2.4	4.3
NO_2	pphm	15	2.6	1.4	1.5	4.9
O_3	pphm	15	1.8	1.1	0.1	2.7
SO_2	pphm	15	1.4	1.1	0.5	3.4

TABLE III-18

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM CONCORD DURING SIX EPISODES 1982-1984

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	18	29	13	10	44
M398MS9	rev/m^3	18	13	6	4	21
ORG98PS9	rev/µg	17	14	11	2	30
ORG98MS9	rev/µg	17	6	5	<1	14
M398NRM	rev/m^3	12	5	2	3	7
NR/98M3	-	12	0.34	0.18	0.15	0.58
BAP	ng/m^3	18	0.3	0.3	0.1	0.8
BKF	ng/m^3	18	0.2	0.1	0.1	0.4
BGP	ng/m^3	18	1.4	0.9	0.5	2.6
COR	ng/m^3	18	0.9	0.4	0.5	1.5
BZO	ng/m^3	18	1.1	1.1	0.2	3.2
ORG	$\mu g/m^3$	17	3.9	2.8	1.5	8.4
TSP	$\mu g/m^3$	18	54	12	38	74
NO_3^-	$\mu g/m^3$	18	7.3	2.8	4.7	12.6
$SO_4^=$	$\mu g/m^3$	18	6.1	1.4	5.0	8.9
BRF	ng/m^3	18	53	31	25	108
PBF	ng/m^3	18	287	132	202	542
BRF/PBF	-	18	0.18	0.08	0.13	0.34
ZNF	ng/m^3	18	20	9	9	31
KF	ng/m^3	18	137	102	76	339
FEF	ng/m^3	18	95	41	58	169
SIF	ng/m^3	18	209	95	134	387
CLF	ng/m^3	18	120	135	27	380
NIF	ng/m^3	18	3	2	2	6
SF	ng/m^3	18	1364	721	641	2614
CO	ppm	18	1.2	0.4	0.5	1.7
NO	pphm	18	2.3	1.3	0.3	4.2
NO_2	pphm	18	3.1	0.9	2.0	4.5
O_3	pphm	18	1.9	0.9	0.3	2.8
SO_2	pphm	18	0.0	0.1	0.0	0.1

TABLE III- 19

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM PITTSBURG DURING SIX EPISODES 1982-1984

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	18	23	9	11	32
M398MS9	rev/m^3	18	10	5	4	17
ORG98PS9	rev/µg	18	11	6	2	18
ORG98MS9	rev/µg	18	5	3	<1	8
M398NRM	rev/m^3	12	4	2	2	5
NR/98M3	-	12	0.31	0.1	0.18	0.39
BAP	ng/m^3	18	0.2	0.1	0.1	0.3
BKF	ng/m^3	18	0.1	0.0	0.1	0.2
BGP	ng/m^3	18	0.8	0.5	0.3	1.7
COR	ng/m^3	18	0.6	0.4	0.2	1.2
BZO	ng/m^3	18	0.7	0.6	0.1	1.4
ORG	µg/m^3	18	3.3	2.5	1.5	6.8
TSP	µg/m^3	18	67	18	52	101
NO_3^-	µg/m^3	18	8.4	5.0	4.5	18.2
$SO_4^=$	µg/m^3	18	7.0	1.3	5.4	8.6
BRF	ng/m^3	18	42	37	22	117
PBF	ng/m^3	18	244	182	109	605
BRF/PBF	-	18	0.17	0.06	0.13	0.27
ZNF	ng/m^3	18	30	7	20	37
KF	ng/m^3	18	162	132	86	429
FEF	ng/m^3	18	153	88	48	256
SIF	ng/m^3	18	337	197	150	610
CLF	ng/m^3	18	188	141	59	393
NIF	ng/m^3	18	5	2	3	9
SF	ng/m^3	18	1437	613	516	2145
CO	ppm	18	0.9	0.4	0.6	1.7
NO	pphm	18	1.3	1.0	0.4	2.8
NO_2	pphm	18	2.2	1.0	0.9	3.7
O_3	pphm	18	2.5	1.3	0.1	4.1
SO_2	pphm	18	0.1	0.1	0.0	0.2

ratios also suggest that Richmond, the furthest west, was exposed least to aged urban pollutants, by the predominantly westerly winds during the periods of sampling.

Individual PAH and total benzene soluble organic (ORG) concentrations were highest at Concord, followed by Richmond. PAH and ORG concentrations were significantly lower than measured in 1981-82. For example, the highest mean BAP levels by station were measured at Concord. The concentration of BAP was 0.3 ng/m^3 in the present study and 0.9 ng/m^3 in the previous study (18).

TSP levels were the highest at Martinez, probably because the sampling site was at ground level (1 m); the other sites were located on the rooftops of one story buildings. Fine fraction soil constituents (FEF, SIF) were also highest at Martinez.

Mean NO_3^- levels showed little geographic variation, ranging from 7.3 $\mu g/m^3$ at Concord to 8.4 $\mu g/m^3$ at Pittsburg. $SO_4^=$ varied from 6.1 $\mu g/m^3$ at Concord to 10.8 $\mu g/m^3$ at Richmond while fine fraction S showed the same geographic variations as $SO_4^=$.

PBF and BRF were both fairly consistent by station. The highest mean PBF was 287 ng/m^3 at Concord, the lowest was 209 ng/m^3 at Richmond. Mean BRF ranged from 53 ng/m^3 at Concord to 42 ng/m^3 at Richmond and Pittsburg. NIF was highest (15 ng/m^3) at the Martinez site, located adjacent to a petrochemical refinery.

CLF was more than four times higher at Richmond (497 $\mu g/m^3$) than at Concord (120 $\mu g/m^3$). The high concentration at Richmond probably reflects its seaward location. It is the most westerly of the four sampling sites.

Among the gaseous pollutants, mean NO was highest at Martinez (3.1 pphm), NO_2 at Concord (3.2 pphm) and O_3 at Pittsburg (2.5 pphm). SO_2 was found primarily at Martinez (1.4 pphm). Mean SO_2 values at the other three stations were $\leq$ 0.1 pphm. SO_2 is derived from local sources at Martinez (e.g. petrochemical refining, sanitary treatment facilities). CO levels (not measured at Martinez) were fairly constant at the other stations.

Correlation Analysis

Correlations between mutagenic density, PAH and selected particulate and gaseous pollutants measured at the four stations are presented in Tables III-20-23. (Complete correlation matrices are provided in the Appendix). Mutagenic density was significantly correlated with NO_3^- at all four stations and was correlated with BRF at three of the stations. However at Concord, mutagenic density was not correlated with either BRF or PBF, despite the fact that mean values of all three variables (mutagenic density, BRF, PBF) were the highest there. PBF and BRF were consistently and highly significantly ($p \leq 0.01$) correlated with COR, at all stations, suggesting further that automotive emissions are significant sources of COR.

The station-by-station comparisons revealed positive associations between mutagenicity and industrial tracers for the first time. Mutagenicity was positively correlated with petrochemical tracers SO_2 and fine fraction sulfur at Richmond and Martinez. At Pittsburg, a highly significant correlation ($p \leq 0.01$) between NIF and both mutagenic density variables ($\pm$S9) was observed (cf Table III-22). NIF was also correlated with each of the three PAH at Pittsburg. These correlations probably reflect co-transport of fine fraction Ni, PAH and mutagens to Pittsburg (typically a receptor site during the six sampling episodes) from upwind sources to the west. The higher nickel concentrations and the lower correlations between nickel and mutagenicity at Martinez provide further evidence for the formation of secondary mutagenic aerosols in the atmosphere.

TABLE III- 20

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS DURING EPISODES AT RICHMOND

	TA98+S9	TA98-S9	BAP	COR	BZO
TA98+S9	1.00	0.93**	0.69**	0.75**	0.87**
TA98-S9	0.93**	1.00	0.72**	0.56*	0.83**
BAP	0.65**	0.72**	1.00	0.26	0.91**
COR	0.75**	0.56*	0.26	1.00	0.55*
BZO	0.87**	0.83**	0.91**	0.55*	1.00
PBF	0.57*	0.36	0.02	0.81**	0.30
BRF	0.80**	0.72**	0.42	0.80**	0.63**
KF	0.52*	0.31	0.09	0.64**	0.15
ZNF	0.31	0.20	-0.03	0.56*	0.30
FEF	0.38	0.18	-0.09	0.64**	0.17
SIF	0.53*	0.27	0.07	0.71*	0.30
CLF	-0.29	-0.24	-0.11	-0.18	-0.23
NIF	0.29	0.16	-0.04	0.21	0.12
SF	0.71**	0.84**	0.72**	0.23	0.73**
NO_3^-	0.70**	0.69**	0.58*	0.37	0.69**
CO	0.66**	0.50*	0.23	0.87**	0.44
NO	0.83**	0.78**	0.67**	0.76**	0.77**
NO_2	0.59**	0.40	0.16	0.67**	0.36
O_3	-0.38	-0.46	-0.67**	0.00	-0.54*
SO_2	0.75**	0.67**	0.73**	0.53*	0.78**

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

TABLE III-21

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS DURING EPISODES AT MARTINEZ

	TA98+S9	TA98-S9	BAP	COR	BZO
TA98+S9	1.00	0.93**	0.29	-0.01	0.49*
TA98-S9	0.93**	1.00	0.33	0.16	0.46
BAP	0.29	0.33	1.00	0.71**	0.83**
COR	-0.01	0.16	0.71**	1.00	0.63**
BZO	0.49*	0.46	0.83**	0.63**	1.00
PBF	0.13	0.33	0.57*	0.84**	0.46
BRF	0.33	0.48*	0.73**	0.85**	0.67*
KF	0.18	0.35	0.56*	0.79**	0.47*
ZNF	0.14	0.34	0.35	0.54*	0.23
FEF	-0.03	0.17	0.43	0.77**	0.28
SIF	-0.16	0.02	0.36	0.73**	0.22
CLF	-0.32	-0.26	-0.25	-0.01	-0.24
NIF	0.10	0.12	-0.23	-0.19	-0.34
SF	0.51*	0.51*	0.04	-0.03	0.19
NO_3^-	0.63**	0.57*	0.16	-0.01	0.47
CO†	-	-	-	-	-
NO	0.55	0.49	0.83**	0.52	0.74*
NO_2	0.29	0.51	0.42	0.74**	0.33
O_3	-0.47	-0.54*	-0.49	-0.37	-0.62*
SO_2	0.74**	0.59*	0.41	-0.02	0.52*

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

†CO not measured.

TABLE III-22

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS DURING EPISODES AT CONCORD

	TA98+S9	TA98-S9	BAP	COR	BZO
TA98+S9	1.00	0.96**	0.34	0.31	0.46
TA98-S9	0.96**	1.00	0.42	0.17	0.49*
BAP	0.34	0.42	1.00	0.18	0.95**
COR	0.31	0.17	0.18	1.00	0.35
BZO	0.46	0.49*	0.95**	0.35	1.00
PBF	-0.14	-0.26	-0.27	0.70**	-0.15
BRF	0.09	-0.05	0.16	0.79**	0.28
KF	0.05	-0.09	0.13	0.74**	0.20
ZNF	-0.13	-0.17	-0.03	-0.04	0.05
FEF	0.00	-0.11	-0.36	0.60**	-0.30
SIF	-0.07	-0.17	-0.12	0.62**	-0.11
CLF	-0.02	0.01	0.55*	-0.06	0.52*
NIF	-0.02	-0.14	0.19	0.77**	0.23
SF	-0.01	-0.07	0.41	0.02	0.46
NO_3^-	0.57*	0.55*	0.71**	0.37	0.82**
CO	0.56*	0.53*	0.04	0.50*	0.15
NO	0.33	0.26	0.21	0.75*	0.36
NO_2	0.04	0.04	-0.26	0.50*	-0.24
O_3	-0.09	-0.11	-0.40	-0.11	-0.39
SO_2	0.38	0.40	0.40	-0.08	0.31

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

TABLE III-23

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS AT PITTSBURG

	TA98+S9	TA98-S9	BAP	COR	BZO
TA98+S9	1.00	0.92**	0.68**	0.63**	0.71**
TA98-S9	0.92**	1.00	0.75**	0.57*	0.71**
BAP	0.68**	0.75**	1.00	0.83**	0.85**
COR	0.63**	0.57*	0.83**	1.00	0.85**
BZO	0.71**	0.71**	0.85**	0.85**	1.00
PBF	0.46	0.41	0.64**	0.82**	0.55*
BRF	0.56*	0.55*	0.73**	0.87**	0.69**
KF	0.49*	0.40	0.50*	0.76**	0.56*
ZNF	0.20	0.31	0.41	0.38	0.35
FEF	0.36	0.41	0.41	0.49*	0.17
SIF	0.17	0.18	0.01	0.13	-0.21
CLF	-0.31	-0.16	-0.07	-0.21	-0.06
NIF	0.73**	0.64**	0.49*	0.57*	0.56*
SF	0.29	0.32	0.25	0.18	0.31
NO_3^-	0.56*	0.61**	0.28	0.20	0.53*
CO	0.71**	0.71**	0.72**	0.78**	0.66**
NO	0.59**	0.51*	0.55*	0.56*	0.42
NO_2	0.25	0.23	0.59*	0.74**	0.49*
O_3	-0.33	-0.50*	-0.39	-0.12	-0.46
SO_2	-0.25	-0.26	-0.32	-0.26	-0.51*

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

To summarize the correlations by station, mutagenic density was correlated positively with source tracers for automotive, secondary and industrial emissions. The strengths of these correlations were different at the four stations. Aerosols collected at Richmond exhibited the strongest association between mutagenicity and automotive tracers (PBF, BRF), Concord showed the strongest association between mutagenicity and secondary aerosols (NO_3^-), while mutagenicity was correlated with petrochemical industrial source tracers at Richmond and Martinez (SO_2, SF). These geographic differences make physical sense in terms of what we know about the sampling stations. Richmond and Martinez have major refineries nearby whereas Concord is located in the Diablo Valley, a downwind receptor area.

4. Episode Comparisons

Although each sampling episode provided only 12 values (or less, due to missing data) for each pollutant variable, comparisons of episodes provide some information about possible seasonal differences in source patterns. Also qualitative comparisons with prevailing meteorological conditions can be made.

Summary Statistics

Summary statistics for air pollutants from each of the six episodes are presented in Table III-24-29. The highest mean mutagenic density (+S9) was 33 rev/m^3, measured during a cold weather episode, (No. VI), in January 1984, and the lowest was 7 rev/m^3, during a warm August 1982 episode (No. I). However, the highest mutagenic specific activity (20 rev/ µg) was measured during September 1983 (No. IV). The lowest ratio of TA98NR/TA98 (0.30) was also observed then suggesting that nitroarenes may have contributed to the high specific activity of aerosols collected in Episode IV.

TABLE III-24

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM EPISODE I
0600 August 23-1800 August 24, 1982

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	4	7	4	3	11
M398MS9	rev/m^3	4	3	1	<1	4
ORG98PS9	rev/µg	4	1.2	<1	<1	1.7
ORG98MS9	rev/µg	4	<1	<1	<1	<1
M398NRM	rev/m^3	-	-	-	-	-
NR/98M3	-	-	-	-	-	-
BAP	ng/m^3	4	0.1	0	0.1	0.1
BKF	ng/m^3	4	0.1	0	0.1	0.1
BGP	ng/m^3	4	0.4	0.1	0.2	0.5
COR	ng/m^3	4	0.4	0.2	0.2	0.5
BZO	ng/m^3	4	0.3	0.1	0.2	0.4
ORG	µg/m^3	4	5.8	0.7	4.8	6.4
TSP	µg/m^3	4	37	11	24	52
NO_3^-	µg/m^3	4	4.1	0.7	3.2	4.7
$SO_4^=$	µg/m^3	4	6.4	0.7	5.7	7.2
BRF	ng/m^3	4	19	6	12	24
PBF	ng/m^3	4	148	58	85	202
BRF/PBF	-	4	0.14	0.01	0.13	0.16
ZNF	ng/m^3	4	24	12	9	37
KF	ng/m^3	4	71	17	50	86
FEF	ng/m^3	4	66	21	39	91
SIF	ng/m^3	4	154	56	91	227
CLF	ng/m^3	4	179	49	132	245
NIF	ng/m^3	4	6	8	2	18
SF	ng/m^3	4	1671	316	1228	1955
CO	ppm	3	0.6	0.2	0.5	0.9
NO	pphm	3	0.5	0.1	0.3	0.6
NO_2	pphm	4	1.9	0.7	1.0	2.6
O_3	pphm	4	2.5	0.2	2.2	2.7
SO_2	pphm	4	0.2	0.2	0.0	0.5

TABLE III- 25

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM EPISODE II
1800 October 12-0600 October 14, 1982

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	4	24	7	16	32
M398MS9	rev/m^3	4	10	2	8	13
ORG98PS9	rev/µg	4	4	3	2	9
ORG98MS9	rev/µg	4	2	1	1	3
M398NRM	rev/m^3	4	4	1	3	5
NR/98M3	-	4	0.43	0.10	0.36	0.58
BAP	ng/m^3	4	0.2	0.1	0.2	0.3
BKF	ng/m^3	4	0.1	0.0	0.1	0.1
BGP	ng/m^3	4	1.5	0.6	0.9	2.2
COR	ng/m^3	4	1.1	0.3	0.7	1.5
BZO	ng/m^3	4	1.1	0.3	0.8	1.4
ORG	µg/m^3	4	7.4	0.7	6.8	8.4
TSP	µg/m^3	4	92	26	69	124
NO_3^-	µg/m^3	4	8.5	1.4	7.5	10.6
$SO_4^=$	µg/m^3	4	6.7	1.1	5.8	7.9
BRF	ng/m^3	4	95	27	56	117
PBF	ng/m^3	4	538	92	407	605
BRF/PBF	-	4	0.17	0.03	0.13	0.20
ZNF	ng/m^3	4	34	11	18	44
KF	ng/m^3	4	350	78	247	429
FEF	ng/m^3	4	243	85	169	357
SIF	ng/m^3	4	512	221	387	843
CLF	ng/m^3	4	101	96	44	244
NIF	ng/m^3	4	12	5	6	17
SF	ng/m^3	4	2025	713	1225	2773
CO	ppm	3	1.5	0.1	1.4	1.7
NO	pphm	3	2.8	1.4	1.4	4.2
NO_2	pphm	4	4.3	0.6	3.7	4.9
O_3	pphm	4	2.4	0.9	1.5	3.5
SO_2	pphm	4	0.3	0.4	0.0	0.9

TABLE III-26

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM EPISODE III
1800 May 17-0600 May 19, 1983

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	4	13	6	7	19
M398MS9	rev/m^3	4	6	2	4	9
ORG98PS9	rev/µg	4	8	4	5	12
ORG98MS9	rev/µg	4	4	1	3	5
M398NRM	rev/m^3	-	-	-	-	-
NR/98M3	-	-	-	-	-	-
BAP	ng/m^3	4	0.1	0.0	0.1	0.2
BKF	ng/m^3	4	0.1	0.0	0.1	0.1
BGP	ng/m^3	4	0.7	0.2	0.5	0.9
COR	ng/m^3	4	0.6	0.1	0.5	0.7
BZO	ng/m^3	4	0.3	0.2	0.1	0.5
ORG	$\mu g/m^3$	4	1.7	0.2	1.5	2.0
TSP	$\mu g/m^3$	4	68	18	47	91
NO_3^-	$\mu g/m^3$	4	6.7	1.0	5.7	8.0
$SO_4^=$	$\mu g/m^3$	4	7.1	1.4	5.3	8.3
BRF	ng/m^3	4	43	9	32	53
PBF	ng/m^3	4	254	16	236	274
BRF/PBF	-	4	0.17	0.05	0.14	0.24
ZNF	ng/m^3	4	31	26	9	68
KF	ng/m^3	4	132	41	76	171
FEF	ng/m^3	4	192	81	101	277
SIF	ng/m^3	4	486	369	147	952
CLF	ng/m^3	4	698	998	62	2173
NIF	ng/m^3	4	6	4	2	12
SF	ng/m^3	4	1040	254	746	1360
CO	ppm	3	1.0	0.4	0.6	1.3
NO	pphm	3	1.1	0.6	0.4	1.4
NO_2	pphm	3	2.8	0.2	2.5	3.0
O_3	pphm	3	3.2	0.7	2.8	4.1
SO_2	pphm	3	0.1	0.1	0.0	0.2

TABLE III-27

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM EPISODE IV
1800 September 12-0600 September 14, 1983

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m	4	25	15	9	44
M398MS9	rev/m	4	12	7	4	21
ORG98PS9	rev/µg	3	20	9	12	30
ORG98MS9	rev/µg	3	9	4	6	14
M398NRM	rev/m	4	2	1	2	3
NR/98M3	-	4	0.30	0.26	0.15	0.69
BAP	ng/m	4	0.1	0.1	0.1	0.2
BKF	ng/m	4	0.1	0.0	0.1	0.1
BGP	ng/m	4	0.5	0.4	0.3	1.1
COR	ng/m	4	0.3	0.3	0.1	0.7
BZO	ng/m	4	0.3	0.3	0.1	0.7
ORG	$\mu g/m^3$	3	1.6	0.1	1.5	1.7
TSP	$\mu g/m^3$	3	62	13	54	77
NO_3^-	$\mu g/m^3$	3	5.7	0.1	5.7	5.8
$SO_4^=$	$\mu g/m^3$	3	6.3	1.8	5.0	8.4
BRF	ng/m	4	23	11	9	32
PBF	ng/m	4	146	67	52	207
BRF/PBF	-	4	0.16	0.02	0.14	0.18
ZNF	ng/m	4	18	9	9	28
KF	ng/m	4	94	29	55	124
FEF	ng/m	4	124	76	26	188
SIF	ng/m	4	292	203	56	487
CLF	ng/m	4	93	90	27	227
NIF	ng/m	4	10	12	2	27
SF	ng/m	4	1414	561	641	1902
CO	ppm	3	1.1	0.2	0.9	1.3
NO	pphm	4	1.8	1.0	0.3	2.5
NO_2	pphm	4	2.0	1.2	0.9	3.3
O_3	pphm	4	2.3	0.5	1.6	2.8
SO_2	pphm	4	0.4	0.6	0.0	1.2

TABLE III-28

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM EPISODE V
1800 October 4-0600 October 6, 1983

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	4	21	7	14	30
M398MS9	rev/m^3	4	9	4	6	15
ORG98PS9	rev/µg	4	12	4	8	17
ORG98MS9	rev/µg	4	6	2	4	8
M398NRM	rev/m^3	4	3	1	3	4
NR/98M3	-	4	0.36	0.06	0.29	0.43
BAP	ng/m^3	4	0.2	0.0	0.1	0.2
BKF	ng/m^3	4	0.1	0.0	0.1	0.2
BGP	ng/m^3	4	1.0	0.4	0.5	1.5
COR	ng/m^3	4	0.6	0.3	0.3	0.9
BZO	ng/m^3	4	0.8	0.2	0.5	1.0
ORG	$\mu g/m^3$	4	1.8	0.2	1.6	1.9
TSP	$\mu g/m^3$	4	57	4	54	63
NO_3^-	$\mu g/m^3$	4	6.5	1.4	4.7	7.7
$SO_4^=$	$\mu g/m^3$	4	9.2	3.2	5.4	13.0
BRF	ng/m^3	4	41	11	28	52
PBF	ng/m^3	4	218	79	137	310
BRF/PBF	-	4	0.21	0.08	0.15	0.33
ZNF	ng/m^3	4	23	5	16	27
KF	ng/m^3	4	91	23	64	120
FEF	ng/m^3	4	97	25	73	120
SIF	ng/m^3	4	162	46	112	202
CLF	ng/m^3	4	171	153	43	393
NIF	ng/m^3	4	6	4	2	12
SF	ng/m^3	4	1097	536	516	1753
CO	ppm	3	1.3	0.5	0.7	1.7
NO	pphm	4	2.2	0.8	1.1	3.0
NO_2	pphm	4	2.6	0.6	2.2	3.5
O_3	pphm	4	2.6	0.1	2.4	2.7
SO_2	pphm	4	0.3	0.6	0.0	1.1

TABLE III-29

SUMMARY STATISTICS FOR AIR POLLUTANTS FROM EPISODE VI
1800 January 4-0600 January 6, 1984

Variable	Units	N	Mean	Standard Deviation	Minimum Value	Maximum Value
M398PS9	rev/m^3	4	33	8	25	43
M398MS9	rev/m^3	4	16	3	12	19
ORG98PS9	rev/µg	4	18	3	16	21
ORG98MS9	rev/µg	4	8	1	7	10
M398NRM	rev/m^3	4	5	1	4	7
NR/98M3	-	4	0.33	0.01	0.32	0.35
BAP	ng/m^3	4	0.4	0.3	0.2	0.8
BKF	ng/m^3	4	0.2	0.1	0.1	0.4
BGP	ng/m^3	4	1.5	0.9	0.6	2.6
COR	ng/m^3	4	0.7	0.3	0.3	1.1
BZO	ng/m^3	4	1.8	1.1	0.9	3.2
ORG	µg/m^3	4	2.2	0.9	1.6	3.5
TSP	µg/m^3	4	66	6	58	73
NO_3^-	µg/m^3	4	15.0	3.4	11.8	18.1
$SO_4^=$	µg/m^3	4	15.0	7.3	8.5	22.2
BRF	ng/m^3	4	52	18	31	67
PBF	ng/m^3	4	150	49	108	202
BRF/PBF	-	4	0.35	0.06	0.28	0.41
ZNF	ng/m^3	4	23	6	17	31
KF	ng/m^3	4	114	22	94	145
FEF	ng/m^3	4	47	8	38	58
SIF	ng/m^3	4	139	17	120	154
CLF	ng/m^3	4	319	45	282	380
NIF	ng/m^3	4	5	1	3	6
SF	ng/m^3	4	3537	1983	2145	6473
CO	ppm	3	1.2	0.2	1.0	1.3
NO	pphm	4	2.7	1.5	0.7	4.3
NO_2	pphm	4	1.9	0.3	1.5	2.3
O_3	pphm	4	0.2	0.1	0.1	0.4
SO_2	pphm	4	1.0	1.6	0.0	3.4

Higher concentrations of total benzene soluble organics were noted in episodes I and II than in episodes III-VI, suggesting a downward trend over time. In contrast, concentrations of specific PAH varied widely from episode to episode. The highest concentrations of PAH were measured in the stagnant October (1982) and cold January (1984) episodes while the lowest PAH concentrations were measured during the warm weather episodes of August 1982 and September 1983.

For many particulate pollutants, the highest concentrations occurred during the October 1982 episode (No. II) (Table III-25). This probably reflects the stagnant conditions which prevailed. (See episode description above.) These pollutants included TSP, PBF, FEF and SIF. An exception was SF, which was highest during the January 1984 episode when easterly transport prevailed.

Previous measurements in Contra Costa County suggested contributions to air pollution from wood burning in winter (18). In the present study the K/Fe ratio associated with airborne particulate matter was used to approximate the impact of wood combustion on ambient concentrations. The K/Fe ratio in soil is approximately 0.5; in emissions from some non-wood combustion sources, the range of ratios found is 0.2 to 0.3. Previously it was shown that the ratio in ambient air containing mostly particles from wood combustion is >8 (44). In the present comparison, the K/Fe ratio ranged from 0.9 to 1.6 in five of the six episodes. However, during January 1984 the K/Fe ratio was higher 2.5. Furthermore the ratio at night was 3.0. This suggests that during the winter episode some of the aerosol was derived from wood combustion, although not a major proportion.

Among the gases, oxides of nitrogen (NO_x) were highest in October 1982 (No. II), O_3 peaked during May 1983 (No. III) and SO_2 varied from a low of 0.1 pphm in May 1983 to a high of 1.1 pphm in January 1984 (No. VI).

Correlation Analysis

Despite the small number of samples points for each episode two-variable correlations were used to help define short-term phenomena. The results are shown in Tables III-30-35. Due to the small sample size, interpretation should be limited.

There was considerable inconsistency from episode to episode of the associations between mutagenic density, on the one hand, and NO_3^-, PBF and BRF, on the other. Positive correlations with PB or BRF were very significant ($p \leq 0.01$) in Episodes I and II, not significant (at the $p \leq 0.05$ level) in No. III, significant in No. IV, and not significant in Episodes V and VI. Mutagenic density and NO_3^- were significantly correlated only in Episode I. Correlations were lowest during episodes when the range of concentrations of the variables was small. When the combined six episode data base was analyzed, the range of concentrations were greater, and mutagenicity was significantly correlated with PBF, BRF and NO_3^-. Thus pollution patterns observed during each short-term episode did not mirror the average pollution pattern observed when the data from six episodes were combined.

Mutagenic density variables (either +S9 or -S9) were correlated with COR in all episodes except No. II. Mutagenicity correlations with BAP and BZO were less frequently observed. Note that during episode No. III, in May 1983, no positive correlations between mutagenic density and any other measured pollutant were observed (cf Table III-32). However, CLF was significantly negatively correlated with mutagenic density ($\pm$S9). Throughout sampling in May the winds were on-shore from the west.

Among the gases, NO_2 was the best correlated with mutagenic density. Significant positive correlations with NO_2 were found in four episodes (No. I, II, IV and V). This association should be investigated further. Finally, CO was correlated with mutagenic density in episodes I (August 1982), and V (October 1984).

TABLE III-30

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS FROM EPISODE I
0600 August 23-1800 August 24, 1982

	TA98+S9	TA98-S9	BAP†	COR	BZO
TA98+S9	1.00	0.90**	0.00	0.52	0.33
TA98-S9	0.90**	1.00	0.00	0.66*	0.33
BAP	0.00	0.00	0.00	0.00	0.00
COR	0.52	0.66*	0.00	1.00	0.59*
BZO	0.32	0.33	0.00	0.59*	1.00
PBF	0.97**	0.88**	0.00	0.56	0.40
BRF	0.87**	0.82**	0.00	0.59*	0.68*
KF	0.29	0.26	0.00	-0.41	-0.17
ZNF	0.83**	0.61*	0.00	0.18	0.04
FEF	0.32	0.03	0.00	-0.26	0.06
SIF	0.20	-0.03	0.00	-0.37	-0.10
CLF	-0.32	-0.43	0.00	-0.49	0.03
NIF	-0.26	-0.46	0.00	-0.49	-0.29
SF	0.29	0.06	0.00	-0.53	-0.38
NO_3^-	0.85**	0.85**	0.00	0.55	0.17
CO	0.28	0.17	0.00	0.44	0.01
NO	0.37	0.17	0.00	0.55	0.23
NO_2	0.89**	0.75**	0.00	0.00	0.14
O_3	0.48	0.38	0.00	0.19	-0.13
SO_2	-0.14	-0.44	0.00	-0.56	-0.45

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

†All values $\leq$ detection limit (0.1ng/m^3)

TABLE III-31

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS FROM EPISODE II
1800 October 12-0600 October 14, 1982

	TA98+S9	TA98-S9	BAP	COR	BZO
TA98+S9	1.00	0.71**	0.59*	0.80**	0.84**
TA98-S9	0.71**	1.00	0.78**	0.53	0.68*
BAP	0.59*	0.78**	1.00	0.71**	0.82**
COR	0.80**	0.53	0.71*	1.00	0.91**
BZO	0.84**	0.68*	0.82**	0.91**	1.00
PBF	0.64*	0.85**	0.77**	0.66*	0.73**
BRF	0.67*	0.84**	0.84**	0.73**	0.80**
KF	0.59*	0.58*	0.62*	0.57	0.69*
ZNF	0.50	0.70*	0.37	0.31	0.40
FEF	0.39	0.75**	0.57	0.27	0.43
SIF	0.13	0.32	0.28	0.15	0.23
CLF	-0.32	0.05	-0.16	-0.39	-0.35
NIF	-0.19	0.16	-0.24	-0.46	-0.40
SF	-0.36	-0.07	-0.38	-0.61*	-0.51
NO_3^-	0.50	0.25	0.10	0.20	0.26
CO	0.82**	0.86**	0.81**	0.80*	0.92**
NO	0.52	0.46	0.56	0.83**	0.70*
NO_2	0.39	0.68*	0.66*	0.53	0.52
O_3	-0.07	-0.53	-0.56	-0.32	-0.33
SO_2	-0.22	-0.07	-0.05	-0.24	-0.13

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

TABLE III-32

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS FROM EPISODE III, 1800 May 17-0600 May 19, 1983

	TA98+S9	TA98-S9	BAP	COR	BZO
TA98+S9	1.00	0.98**	-0.37	0.46	-0.24
TA98-S9	0.98**	1.00	-0.33	0.49	-0.17
BAP	-0.37	-0.33	1.00	0.21	0.72**
COR	0.46	0.49	0.21	1.00	0.56
BZO	-0.24	-0.17	0.72**	0.56	1.00
PBF	0.44	0.47	0.28	0.81**	0.67*
BRF	0.06	0.04	0.28	0.66*	0.58*
KF	-0.38	-0.32	0.41	0.04	0.47
ZNF	-0.03	0.02	0.16	0.41	0.55
FEF	-0.01	0.07	0.62*	0.09	0.57
SIF	-0.22	-0.18	0.70*	-0.17	0.45
CLF	-0.66*	-0.73**	-0.17	-0.33	-0.17
NIF	-0.41	-0.30	0.49	0.10	0.79**
SF	-0.40	-0.33	0.70*	0.03	0.68*
NO_3^-	0.15	0.26	0.40	0.49	0.61*
CO	-0.03	-0.06	0.00	0.70*	0.63
NO	0.03	0.06	0.00	0.83**	0.70*
NO_2	0.40	0.45	0.00	0.73*	0.78*
O_3	0.19	0.25	0.00	-0.18	-0.11
SO_2	0.34	0.38	0.00	0.20	0.43

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

TABLE III-33

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS FROM EPISODE IV
1800 September 12-0600 September 14, 1983

	TA98+S9	TA98-S9	BAP	COR	BZO
TA98+S9	1.0	0.97**	0.61*	0.68*	0.70*
TA98-S9	0.97**	1.00	0.62*	0.65*	0.74**
BAP	0.61*	0.62*	1.00	0.86**	0.89**
COR	0.68*	0.65*	0.86**	1.00	0.80**
BZO	0.70*	0.74**	0.89**	0.80**	1.00
PBF	0.68*	0.70*	0.63*	0.41	0.63*
BRF	0.51	0.56	0.48	0.26	0.52
KF	0.40	0.48	0.06	0.01	0.06
ZNF	0.28	0.29	-0.21	-0.31	-0.24
FEF	0.37	0.41	-0.06	-0.19	-0.02
SIF	0.25	0.29	-0.19	-0.33	-0.17
CLF	-0.31	-0.25	0.21	-0.15	-0.09
NIF	-0.12	-0.09	-0.39	-0.53	-0.10
SF	-0.54	-0.48	-0.56	-0.70*	-0.49
NO_3^-	0.33	0.38	-0.15	0.03	0.14
CO	0.52	0.54	0.35	0.58	0.45
NO	0.47	0.39	0.00	0.06	0.09
NO_2	0.57	0.60*	0.58*	0.47	0.82**
O_3	0.10	0.13	-0.45	-0.30	-0.35
SO_2	0.02	0.06	-0.29	-0.42	-0.02

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

TABLE III-34

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS FROM EPISODE V
1800 October 4-0600 October 6, 1983

	TA98+S9	TA98-S9	BAP	COR	BZO
TA98+S9	1.00	0.96**	0.62*	0.79**	0.83**
TA98-S9	0.96**	1.00	0.51	0.64*	0.70*
BAP	0.62*	0.51	1.00	0.61*	0.62*
COR	0.79**	0.64*	0.61*	1.00	0.94**
BZO	0.83**	0.70*	0.62*	0.94**	1.00
PBF	0.50	0.41	0.30	0.62*	0.66*
BRF	0.27	0.18	0.25	0.51	0.56
KF	0.13	0.03	0.16	0.50	0.48
ZNF	0.61*	0.55	0.21	0.65*	0.81**
FEF	-0.02	-0.04	0.06	0.03	0.24
SIF	-0.04	-0.02	0.09	-0.02	0.22
CLF	-0.50	-0.39	-0.31	-0.45	-0.47
NIF	-0.25	-0.29	0.13	-0.14	0.04
SF	0.14	0.03	0.09	0.53	0.40
NO_3^-	0.29	0.30	-0.07	0.05	0.14
CO	0.81**	0.70*	0.51	0.83**	0.71*
NO	0.61*	0.54	0.24	0.57	0.65*
NO_2	0.79**	0.81**	0.68*	0.45	0.54
O_3	0.04	0.06	-0.40	0.11	0.12
SO_2	-0.51	-0.49	-0.23	-0.53	-0.43

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

TABLE III- 35

CORRELATIONS BETWEEN MUTAGENIC DENSITY (rev/m^3), SELECTED PAH AND AIR POLLUTANTS FROM EPISODE VI
1800 January 4-0600 January 6, 1984

	TA98+S9	TA98-S9	BAP	COR	BZO
TA98+S9	1.00	0.91**	0.31	0.85**	0.48
TA98-S9	0.91**	1.00	0.39	0.81**	0.50
BAP	0.31	0.39	1.00	0.54	0.96**
COR	0.85**	0.81**	0.54	1.00	0.67*
BZO	0.48	0.50	0.96**	0.67*	1.00
PBF	0.53	0.40	0.18	0.60*	0.25
BRF	0.36	0.24	0.31	0.46	0.33
KF	0.10	-0.03	0.22	0.15	0.20
ZNF	-0.31	-0.26	-0.23	-0.26	-0.30
FEF	0.27	0.13	-0.10	0.26	0.01
SIF	-0.03	-0.05	0.04	0.03	-0.01
CLF	-0.34	-0.48	0.27	-0.17	0.18
NIF	-0.06	-0.10	-0.27	-0.41	-0.24
SF	0.04	-0.00	-0.06	0.05	-0.04
NO_3^-	-0.14	-0.07	-0.40	-0.56	-0.40
CO	0.44	0.51	0.21	0.60	0.24
NO	0.03	-0.01	0.27	0.08	0.20
NO_2	0.40	0.29	0.52	0.50	0.57
O_3	0.53	0.51	-0.13	0.45	0.01
SO_2	-0.32	0.38	-0.29	-0.58*	-0.41

*Significant at the $p \leq 0.05$ level.

**Significant at the $p \leq 0.01$ level.

D. Conclusions

An effective strategy to control levels of mutagenic density and PAH in community aerosols should be guided by an understanding of the primary sources and secondary transformations which produce the mutagens and PAH. Our analysis has shown that the aerosol variables which are the best predictors of mutagenic density are NO_3^- and PBF or BRF. Furthermore, the predictive value of NO_3^- is area-wide. Thus mutagens in particulate matter behaved like both primary automotive emission products and secondary aerosols. The diurnal differences in predictive value of PBF may be the result of meteorological effects. During the daytime, ventilation was generally good so contributions of area-wide secondary pollution were predominant. During the nighttime, lower inversions and lighter surface winds presumably unmasked local transportation sources. The association of mutagenicity with NO_3^--containing aerosols could also be related to atmospheric (or filter) transformations of mutagens catalyzed by HNO_3. Mutagenic density was also correlated with NO and NO_2. These correlations were higher at night than by day, especially with NO_2. Nitration reactions of PAH involving NO_2 and NO_3^- radical at night have recently been suggested by Pitts et al (23).

Among the PAH, coronene was strongly associated with automotive tracers (PBF, BRF) but not with NO_3^-. Benzanthrone, a partially oxidized carbonyl-arene, behaved more like mutagenic density than COR. That is, BZO was associated with NO_3^-, as well as with PBF and BRF.

Geographic comparisons revealed differences in associations between automotive tracers and mutagens at different stations. Correlations between mutagenic density and automotive tracers (PBF and BRF) were highest at Richmond and Pittsburg and lowest at Concord.

A positive correlation between mutagenic density and NIF was observed at Pittsburg, but not at the other three locations. It should be noted that Pittsburg site was generally a receptor site (downwind and to the east of the refineries) during episode sampling. Martinez, which is closest to the refineries, had the

highest average nickel concentrations but the lowest average mutagenicity. This suggests that the refinery emissions are not identified with primary mutagenic aerosol emissions but may contribute to secondary mutagenic aerosol concentrations at downwind locations.

Mutagenicity was also correlated with S compounds (SF, SO_2) at Richmond and Martinez, both industrial centers. Thus sulfur producing sources, including some industries, may also contribute to mutagenic aerosols. Major industrial sources of sulfur oxides are refineries in Richmond (Chevron), Martinez (Shell, Tosco) and Benicia (Exxon) and a chemical plant in Rodeo (Union) (28).

CHAPTER IV.
SEASONAL VARIATIONS AND TRENDS IN THE CONCENTRATIONS OF MUTAGENS, PAH AND STANDARD PARTICULATE POLLUTANTS IN CONTRA COSTA COMMUNITY AEROSOLS

A. Introduction

The results of chronic monitoring studies provide critical baseline information against which the impact of new or expanding technologies (e.g. diesel cars, waste-to-energy conversions) can be measured. Although intensive sampling is required for source identification (Chapter III), a chronic monitoring strategy is essential to identify trends in the levels of toxic air contaminants.

In our previous CARB-sponsored project in Contra Costa County (18), large seasonal variations in PAH concentrations were observed. Concentrations were about five times higher in winter than in spring. Qualitatively similar but smaller seasonal swings were exhibited by mutagenic density, total mass, lead and other particulate pollutants. We concluded that these seasonal patterns resulted primarily from meteorological variations, not seasonal source differences. However, we also suggested that wood smoke from fireplaces during the winter contributed significantly to PAH but not to mutagenic aerosol concentrations. In the Bay Area, seasonal changes in dispersal of pollutants are due to changes in wind direction from west to east, wind speeds and inversion heights. Higher concentrations of particulate pollutants during winter are generally observed.

In the previous study, we also concluded that annual average mutagenic density and PAH concentrations in Contra Costa County had not changed significantly between 1979 and 1982. The present study extends the analysis of seasonal variations and trends through June, 1984 using the same logistical plan (Figure I-2).

B. Experimental Methods

Hi-vol samples were collected every sixth day at Concord, Richmond and Pittsburg and used to prepare composite samples for Ames and PAH testing. Locations

and descriptions of the sites are found in Chapter III above. Other particulate pollutants analyzed in the composites were TSP, LEAD, $SO_4^=$, NO_3^- and ORG. A portion of each filter was composited for PAH and mutagenicity testing. (Prior to compositing, filters were stored for up to 2 years at -10°C in the dark). Separate composites were prepared for each station. Filters from each of the three stations were composited over four-month intervals (July-October, November-February, March-June), to give composite samples for analysis. These periods approximate the three meteorological seasons in the San Francisco Bay air basin and also corresponds with those used in previous studies in Contra Costa County (6,18).

In the current project, samples collected during the period July 1982-June 1984 were composited for analysis of PAH and mutagenic activity. Analysis of these samples provides a continuous data base of concentrations of specific PAH and mutagenic activity found in Contra Costa air particulate material collected over a 60 month period, from November 1979 through October 1984. Results of PAH and mutagenicity measurements in composite samples were compared with other particulate matter pollutants on a season-by-season and annual basis. The PAH and mutagenicity levels were also compared with those measured previously in Contra Costa County and elsewhere.

Air particulate material for mutagenic and PAH testing was collected on 8" x 10" glass fiber filters (Whatman) in standard hi-vol samplers. The sampling rate was 55-60 m^3 per hour.

Analyses of the standard chemical pollutants measured in the ARB air quality network were carried out by the BAAQMD and AIHL using the standard methods. TSP is determined gravimetrically, Pb by energy dispersive x-ray fluorescence, $SO_4^=$ turbidimetrically, by SulfaVer, NO_3^- by a colorimetric procedure utilizing NitraVer 6 and NitraVer 3 pillows and ORGANICS by benzene extraction followed by gravimetric determination (Table I-2) (28,31).

Compositing for mutagenic and PAH testing was performed by cutting pieces from each filter, combining filter disks and extracting with "trisolvent" as described above. To measure mutagenicity of composites, the standard Ames Salmonella/mammalian microsome test was used as described in Chapter III. Methods for the analysis of selected PAH (BAP, BKF, BGP, COR, BZO) employed HPLC with ultraviolet and fluorescence detection and were also as previously decribed (18).

C. Results and Discussion

All results of composite sample analysis are listed in Appendix IV.

Comparison by Station

Mean concentrations for pollutants measured at each station are presented in Table IV-1. Major station-to-station differences were not apparent for most variables, including mutagenic density. Among the PAH there were exceptions however. Concentrations of BAP, BGP, COR and BZO were about twice as high at Concord as at Pittsburg. Total benzene soluble organics (ORG) and lead were also the highest at Concord.

Over the 60 months of composite sampling, Richmond had the highest mutagenic density (11.4 rev/m^3, +S9) and Pittsburg the lowest (10.0 rev/m^3, +S9). Mutagenic densities with metabolic activation (+S9) were about twice those measured without it (-S9) at all three stations. Thus the relative amounts of indirect and direct-acting mutagens were about the same at all locations. Richmond experienced the highest $SO_4^=$ levels (7.4 $\mu g/m^3$), but the lowest NO_3^- pollution levels (4.8 $\mu g/m^3$). Petrochemical refining probably contributed to the $SO_4^=$ at Richmond. As noted above, refineries located in Richmond are major point sources of sulfur oxides. The largest fraction of sulfur oxides released by burning fossil fuels is SO_2. $SO_4^=$ is considered a secondary pollutant, except from sea salt and surface entrainment. However, a proportion (1-2%) of the sulfur oxides from fossil fuel combustion is released as primary $SO_4^=$ (46).

Seasonal Variations

The seasonal variations are shown in Table IV-2. The November-February (winter) season had the highest concentrations for all the pollutants measured except

TABLE IV-1

MEAN AND STANDARD DERIVATIONS IN CONCENTRATIONS
OF AIR POLLUTANTS SAMPLED AT THREE CONTRA COSTA STATIONS:
NOVEMBER 1979-OCTOBER 1984

			Station					
			Richmond		Concord		Pittsburg	
Variable	Units	N	Mean	S.D.	Mean	S.D.	Mean	S.D.
TA98P	rev/m^3	15	11	(7)	11	(7)	10	(7)
TA98M	rev/m^3	15	6	(3)	6	(3)	6	(4)
TA98NRP	rev/m^3	15	5	(3)	6	(3)	5	(3)
TA98NRM	rev/m^3	15	3	(2)	2	(1)	2	(1)
TA98NRM/TA98M	-	15	0.42	(0.22)	0.40	(0.30)	0.43	(0.34)
BAP	ng/m^3	15	0.3	(0.3)	0.6	(0.8)	0.2	(0.2)
BKF	ng/m^3	15	0.2	(0.2)	0.3	(0.3)	0.2	(0.2)
BGP	ng/m^3	15	1.5	(1.3)	2.1	(2.1)	1.1	(1.0)
COR	ng/m^3	15	1.0	(0.9)	1.2	(1.2)	0.7	(0.6)
BZO	ng/m^3	15	0.5	(0.6)	1.1	(1.5)	0.6	(0.8)
ORG	µg/m^3	15	3.7	(1.5)	5.1	(3.9)	3.4	(1.6)
MASS (TSP)	µg/m^3	15	57	(9)	51	(15)	64	(13)
LEAD (Hi Vol)	µg/m^3	15	0.27	(0.12)	0.34	(0.18)	0.23	(0.10)
NO_3^-	µg/m^3	14*	4.8	(2.4)	6.0	(2.5)	6.6	(2.9)
$SO_4^=$	µg/m^3	14*	7.4	(1.7)	5.2	(1.3)	6.6	(1.5)

*Data from July-October 1984 missing.

S.D. = Standard Deviation

TABLE IV-2

SEASONAL VARIATIONS IN CONTRA COSTA
AIR POLLUTANT CONCENTRATIONS (THREE STATION AVERAGES):
NOVEMBER 1979-JUNE 1984

			Station					
			Nov-Feb		March-June		July-Oct	
Variable	Units	N	Mean	S.D.	Mean	S.D.	Mean	S.D.
TA98P	rev/m^3	15	14	(8)	8	(6)	11	(5)
TA98M	rev/m^3	15	7	(4)	5	(3)	5	(2)
TA98NRP	rev/m^3	15	8	(2)	3	(2)	5	(2)
TA98NRM	rev/m^3	15	3	(1)	2	(1)	2	(1)
TA98NRM/TA98M	-	15	0.53	(0.33)	0.39	(0.27)	0.33	(0.14)
BAP	ng/m^3	15	0.9	(0.6)	0.1	(0.02)	0.1	(0.03)
BKF	ng/m^3	15	0.5	(0.2)	0.1	(0.04)	0.1	(0.03)
BGP	ng/m^3	15	3.5	(1.5)	0.6	(0.3)	0.7	(0.2)
COR	ng/m^3	15	1.8	(1.0)	0.5	(0.3)	0.5)	(0.3)
BZO	ng/m^3	15	1.9	(1.1)	0.1	(0.1)	0.2	(0.1)
ORG	µg/m^3	15	6.9	(2.5)	2.4	(0.9)	2.8	(1.0)
MASS (TSP)	µg/m^3	15	64	(13)	50	(10)	58	(13)
LEAD (Hi Vol)	µg/m^3	15	0.42	(0.15)	0.19	(0.04)	0.22	(.10)
NO_3^-	µg/m^3	15	8.6	(2.2)	3.6	(0.8)	*5.0	(1.2)
$SO_4^=$	µg/m^3	15	6.8	(2.2)	5.7	(1.1)	*6.8	(1.3)

*N=14, missing July-Oct. 1984.

S.D. = Standard Deviation

$SO_4^=$. Levels of $SO_4^=$ were the same during the July-October and November-February seasons (i.e. 6.8 $\mu g/m^3$) and only about twenty percent lower during March-June (5.7 $\mu g/m^3$).

Concentrations of mutagenic density, Pb, NO_3^- and ORG were all about twice as high in the winter as in the spring (March-June).

In confirmation of earlier results (18), levels of specific PAH showed the largest seasonal variations. The concentration of BAP was 0.9 ng/m^3 in November-February and $\leq$0.1 ng/m^3 during the other two seasons. BKF, BGP and COR were all 4-6 times more concentrated in winter while BZO was more than 10 times higher in the winter season. The large seasonal changes among the PAH could result (partially) from differences in source patterns. Residential wood combustion would be expected to contribute more to PAH pollution in the winter. Seasonal variations may also reflect selective losses of PAH in warmer seasons through chemical tranformations in the atmosphere or through volatilization or chemical transformations during sampling on filters. These are topics for other studies.

With respect to possible atmospheric formation of nitroarenes, we note that the TA98NR/TA98 ratio was lower in the warm weather seasons (March-June (0.39) and July-October (0.36)) than in winter (November-February (0.53)). The lower the ratio, the greater the fraction of mutagenic activity contributed by nitro-organics, including some NO_2 PAH. Regarding TA98NR, some caveats should be included. Strain TA98NR is deficient in the bacterial nitroreductase which catalyzes the activation of most mononitroarenes (e.g. 1-nitropyrene) to mutagens. Thus, a lower response in TA98NR relative to TA98 probably indicates the presence of mononitroarenes in the sample. However, certain highly mutagenic dinitroarenes (e.g. 1,8 dinitropyrene) are activated by a different nitroreductase, which is functional in TA98NR. Since dinitropyrenes are highly mutagenic in both TA98 and TA98NR, the ratio of TA98NR/TA98 could be high yet the sample could contain these compounds and be highly mutagenic. (Another nitroreductase-deficient strain, TA98/1,8-DNP_6, which lacks the specific nitro reductase required for dinitropyrene activation can be used to indicate the presence of dinitropyrenes in samples.) (47).

The observation that higher concentrations of PAH, mutagenic density and other particulate matter pollutants occur in winter is consistent with results of our earlier study in Contra Costa County (18). Values of mutagenic density are also comparable to, albiet somewhat lower than those measured in urban and residential areas in Los Angeles (23) and elsewere (16,48).

Trends

All data used in the analysis of trends are included in Appendix IV.

As described in the following, one of the most interesting and puzzling results of this research is the apparent downward trend in some aerosol pollutant concentrations and the apparent increasing trend in mutagenic density over time.

Despite seasonal variations, two standard measures of particulate matter pollution (Pb, NO_3^-) showed overall downward trends during the period (Figures IV-1-2). TSP and $SO_4^=$ levels were fairly constant (Figures IV-3-4). Similar trends were reported in our earlier study. It is perhaps relevant to note that some of this study was conducted during some of the wettest years ever recorded in California.

On an annual basis PAH (and ORG) concentrations were fairly constant over time; the exception was in one unusually high winter season (November 1982-February 1983) (Figures IV-5-8). The explanation for this one season excursion was not obviously related to average meteorology during the four months of sampling (38). November was cooler, windier and much wetter than normal, December had nearly normal precipitation and ventilation, January was dry and stagnant in the first half and wet and windy in the second half, while February's weather was dominated by rain.

Quantitative comparisons of trends in the inorganic and organic aerosol pollutants described above are shown in Appendix V. Linear regression analysis demonstrated that between 1979 and 1984, statistically significant ($p \leq 0.05$) decreases in Pb concentrations occurred during the Nov.-Feb. and July-Oct. seasons, as well as

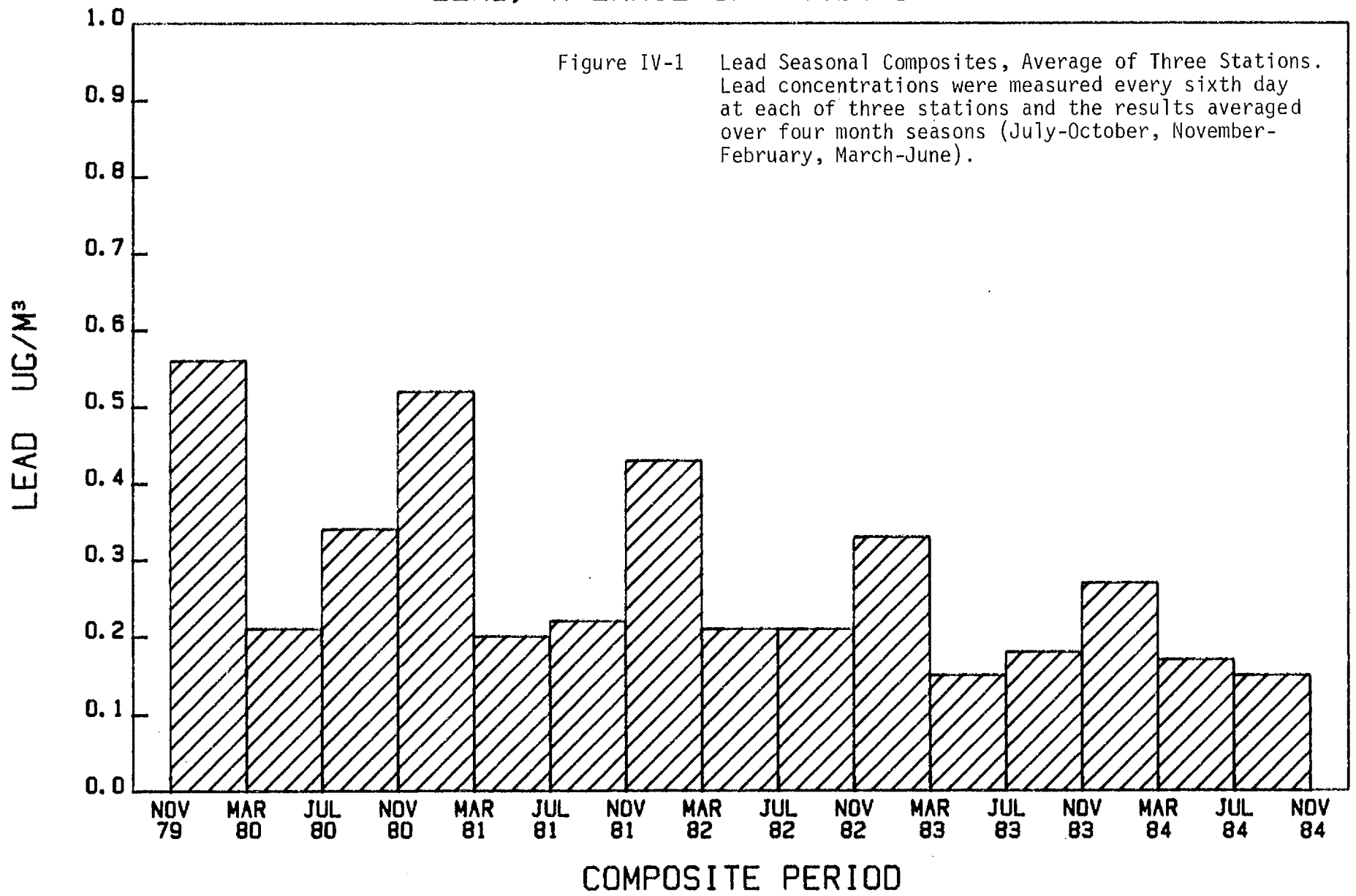


Figure IV-1 Lead Seasonal Composites, Average of Three Stations. Lead concentrations were measured every sixth day at each of three stations and the results averaged over four month seasons (July-October, November-February, March-June).

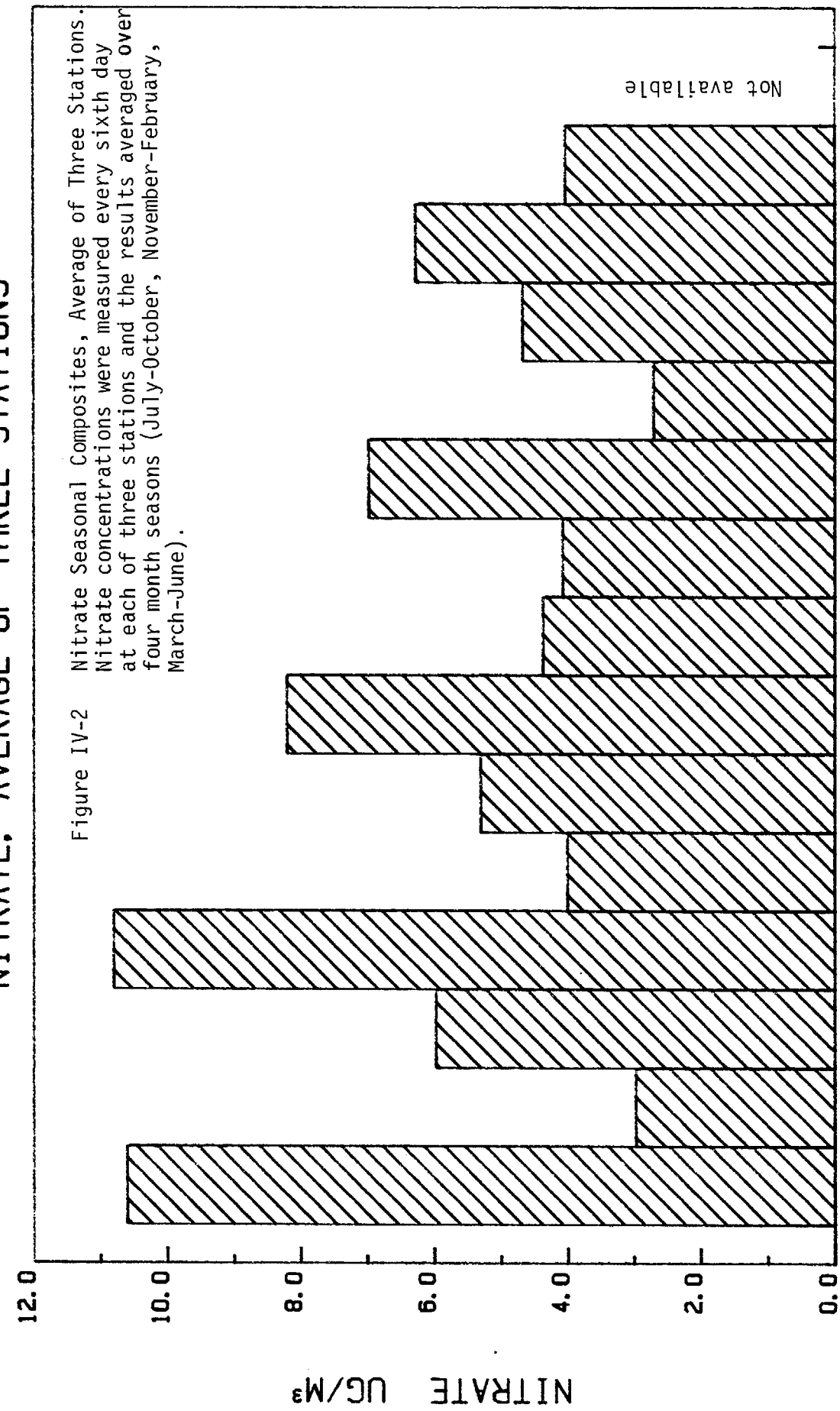


Figure IV-2 Nitrate Seasonal Composites, Average of Three Stations. Nitrate concentrations were measured every sixth day at each of three stations and the results averaged over four month seasons (July-October, November-February, March-June).

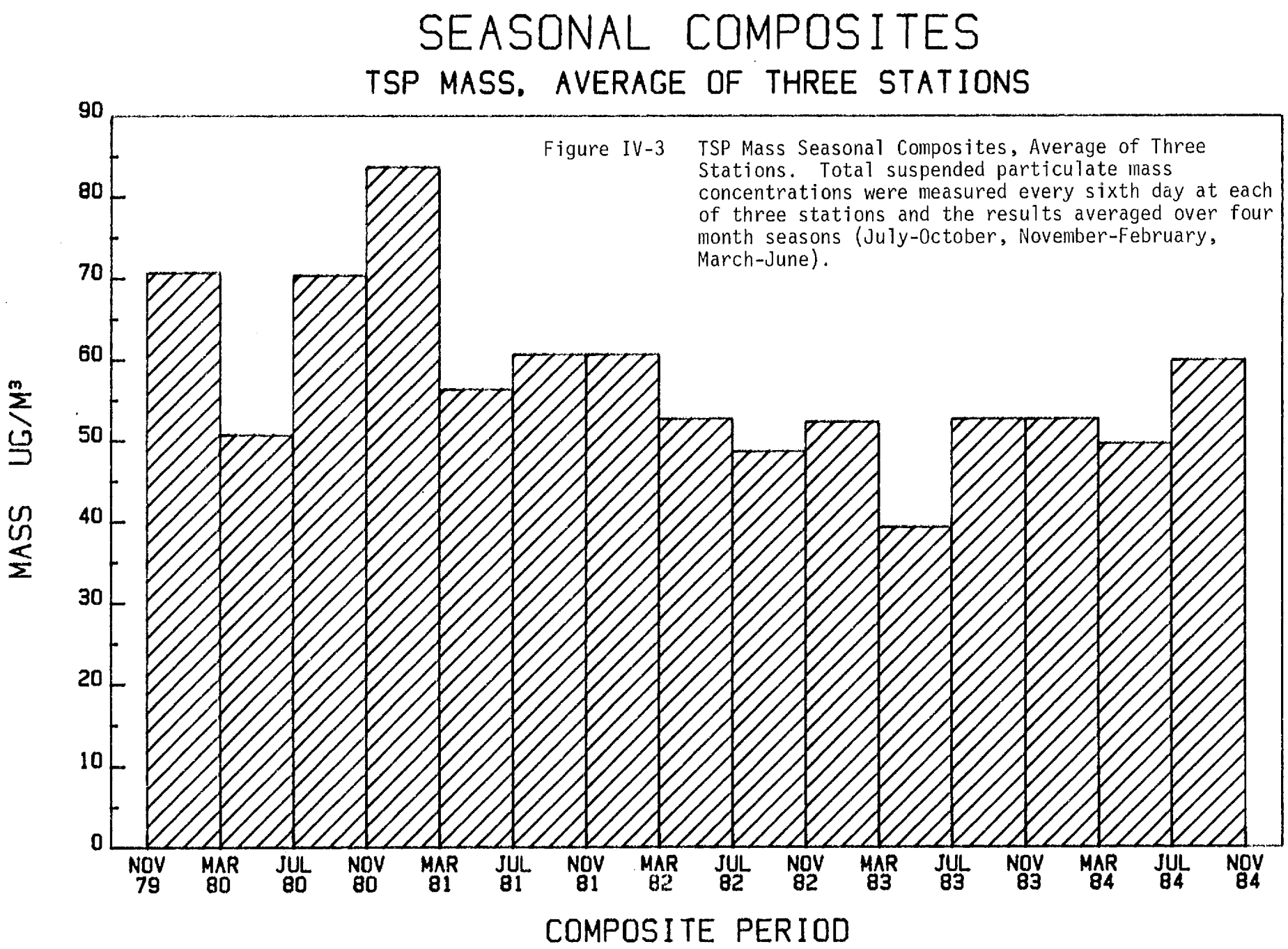


Figure IV-3 TSP Mass Seasonal Composites, Average of Three Stations. Total suspended particulate mass concentrations were measured every sixth day at each of three stations and the results averaged over four month seasons (July-October, November-February, March-June).

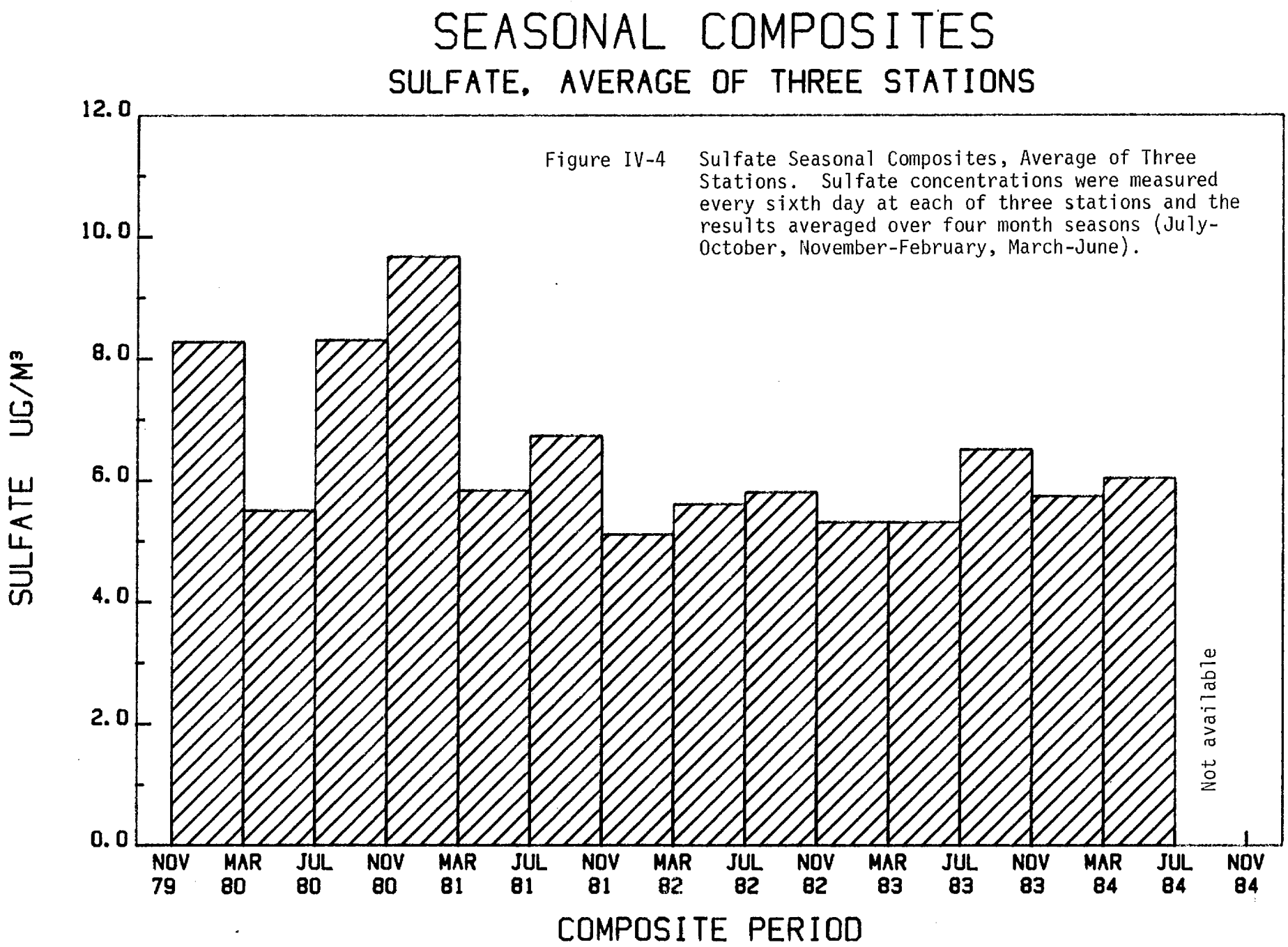


Figure IV-4 Sulfate Seasonal Composites, Average of Three Stations. Sulfate concentrations were measured every sixth day at each of three stations and the results averaged over four month seasons (July-October, November-February, March-June).

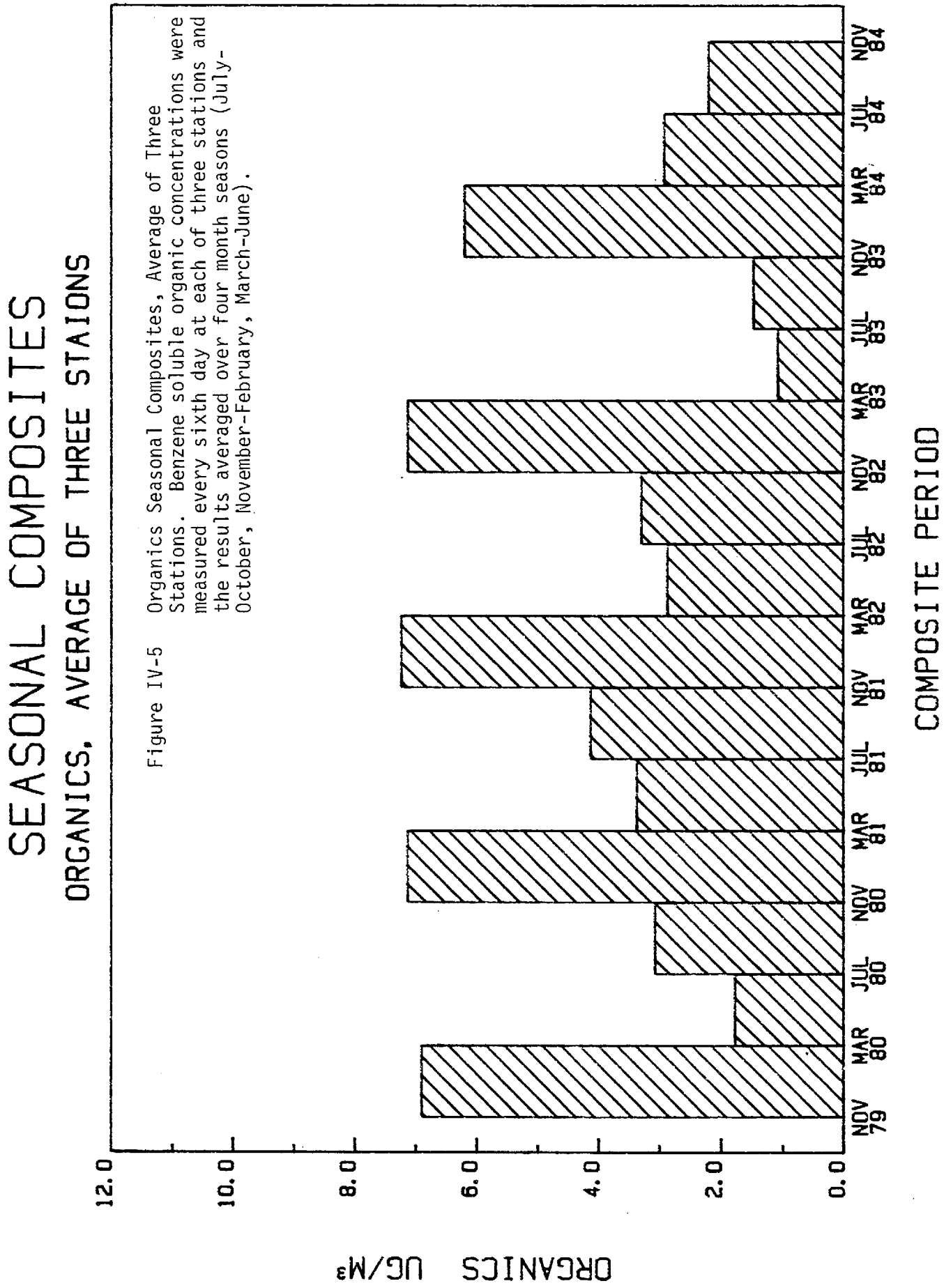


Figure IV-5 Organics Seasonal Composites, Average of Three Stations. Benzene soluble organic concentrations were measured every sixth day at each of three stations and the results averaged over four month seasons (July-October, November-February, March-June).

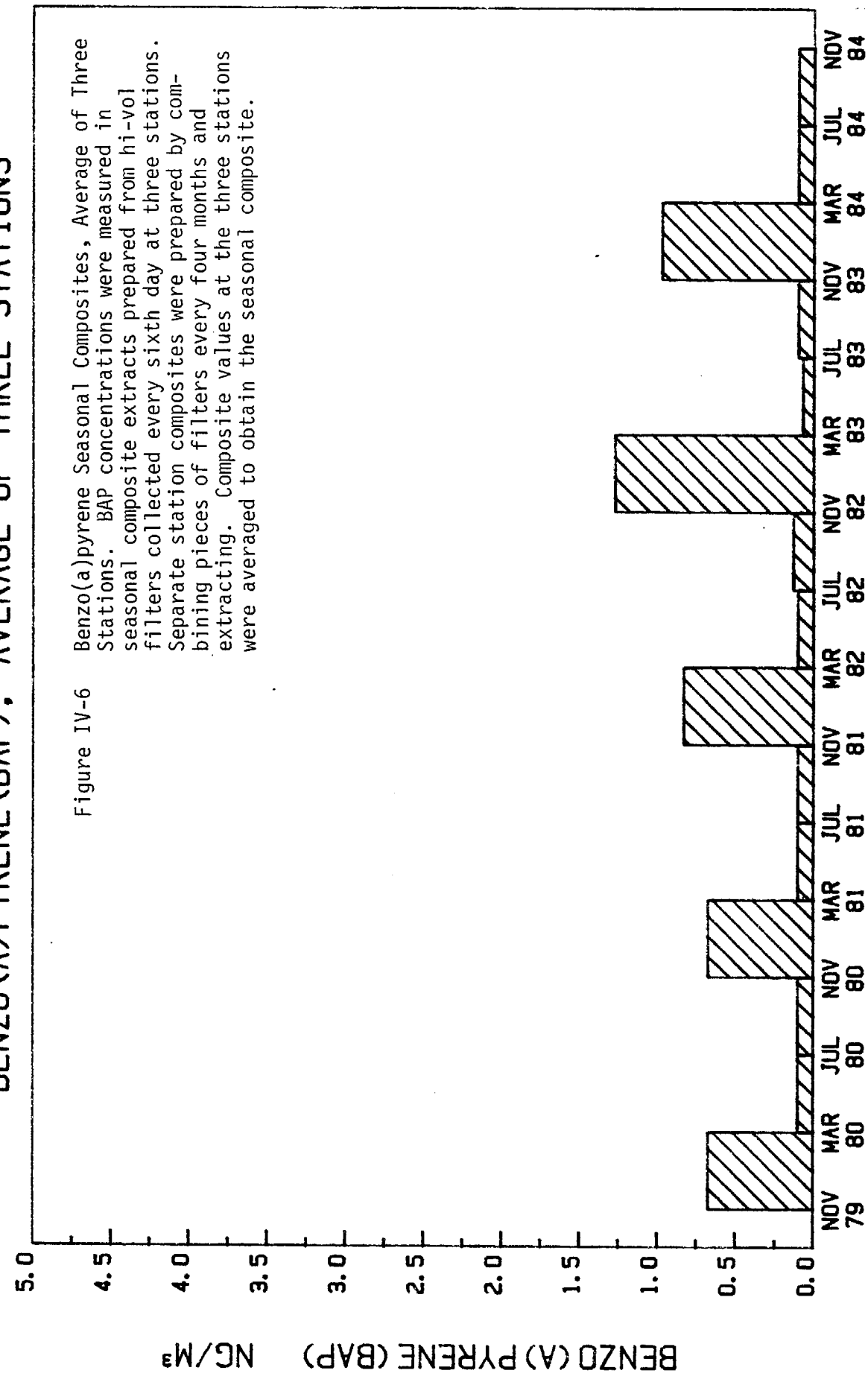


Figure IV-6 Benzo(a)pyrene Seasonal Composites, Average of Three Stations. BAP concentrations were measured in seasonal composite extracts prepared from hi-vol filters collected every sixth day at three stations. Separate station composites were prepared by combining pieces of filters every four months and extracting. Composite values at the three stations were averaged to obtain the seasonal composite.

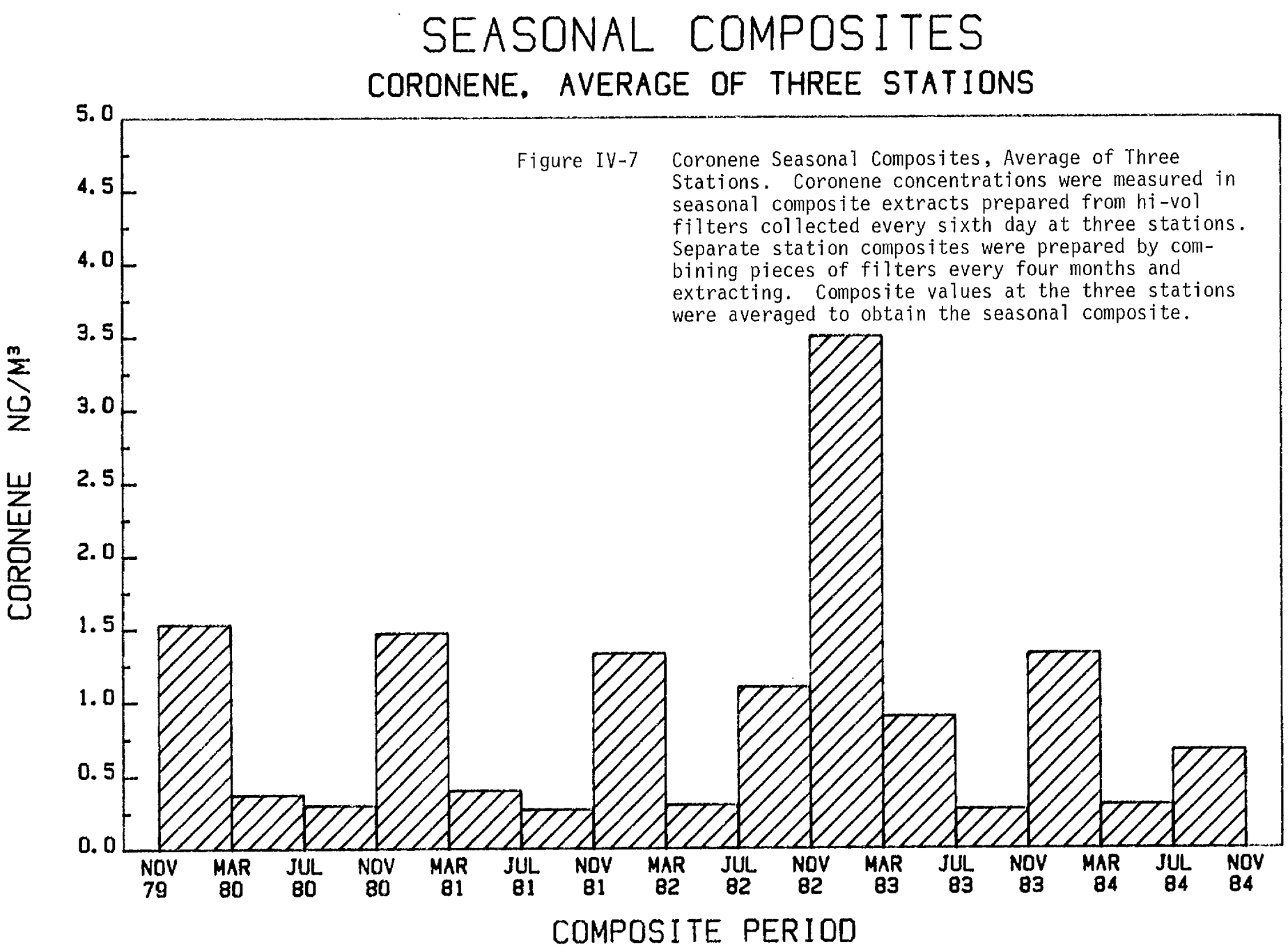


Figure IV-7 Coronene Seasonal Composites, Average of Three Stations. Coronene concentrations were measured in seasonal composite extracts prepared from hi-vol filters collected every sixth day at three stations. Separate station composites were prepared by combining pieces of filters every four months and extracting. Composite values at the three stations were averaged to obtain the seasonal composite.

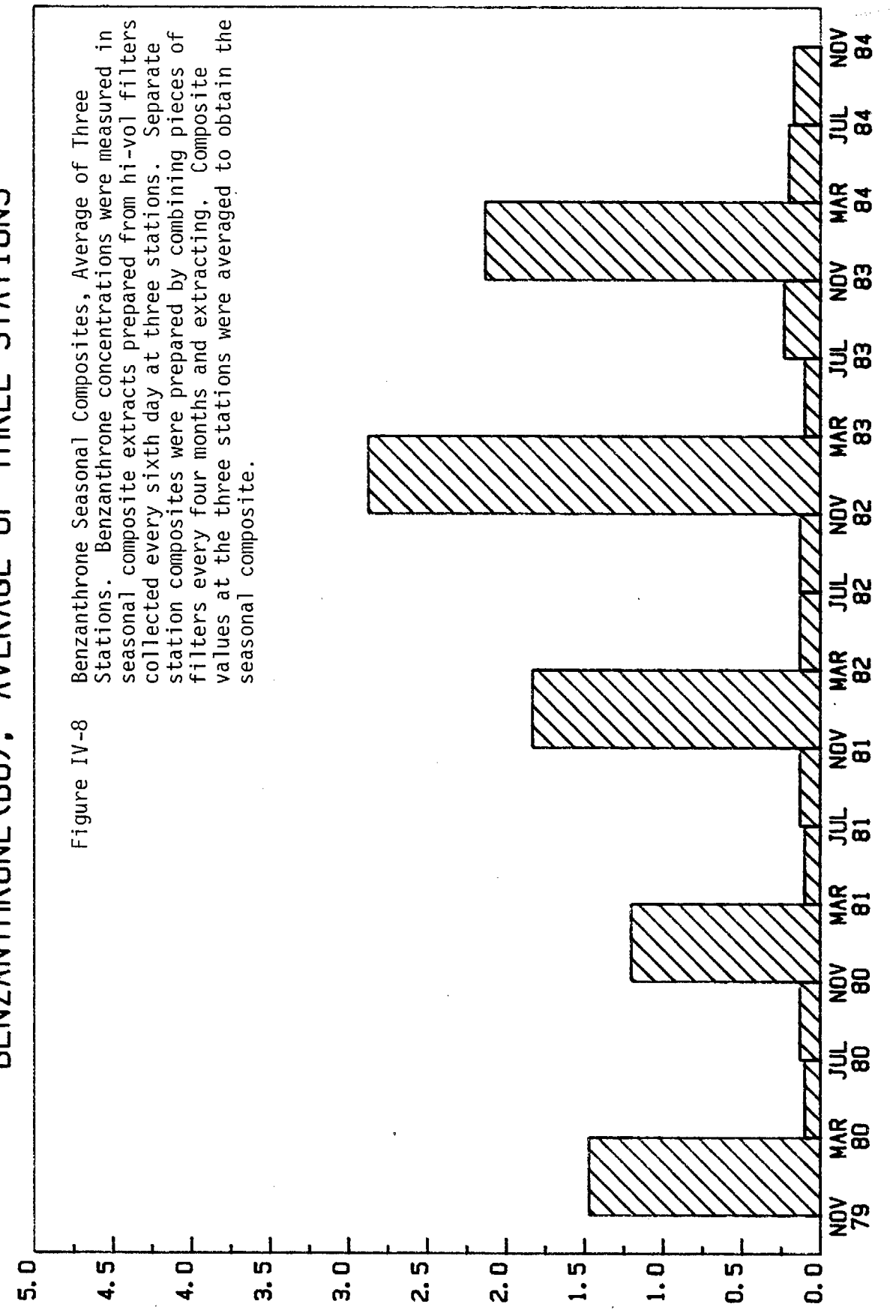


Figure IV-8 Benzanthrone Seasonal Composites, Average of Three Stations. Benzanthrone concentrations were measured in seasonal composite extracts prepared from hi-vol filters collected every sixth day at three stations. Separate station composites were prepared by combining pieces of filters every four months and extracting. Composite values at the three stations were averaged to obtain the seasonal composite.

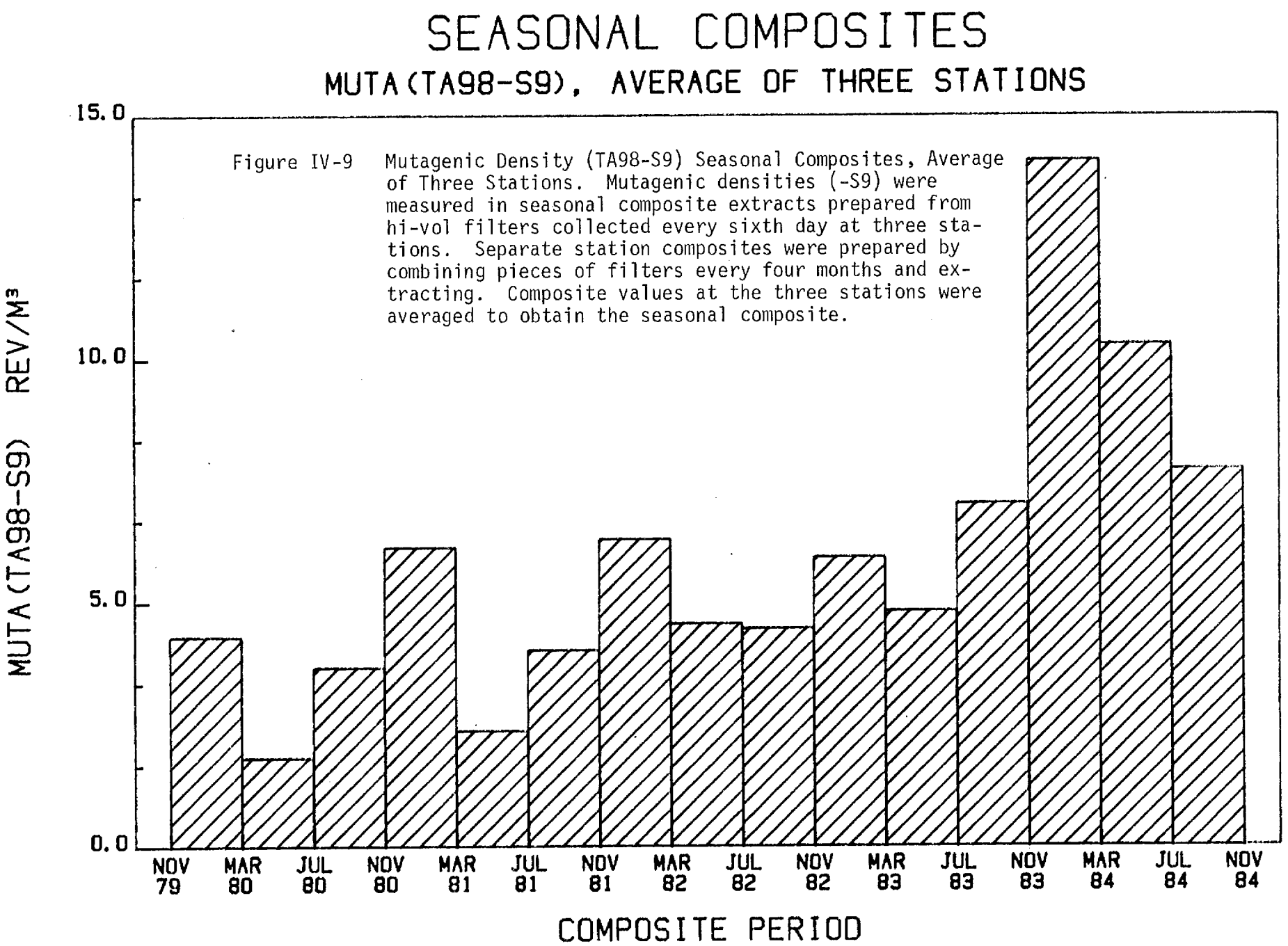


Figure IV-9 Mutagenic Density (TA98-S9) Seasonal Composites, Average of Three Stations. Mutagenic densities (-S9) were measured in seasonal composite extracts prepared from hi-vol filters collected every sixth day at three stations. Separate station composites were prepared by combining pieces of filters every four months and extracting. Composite values at the three stations were averaged to obtain the seasonal composite.

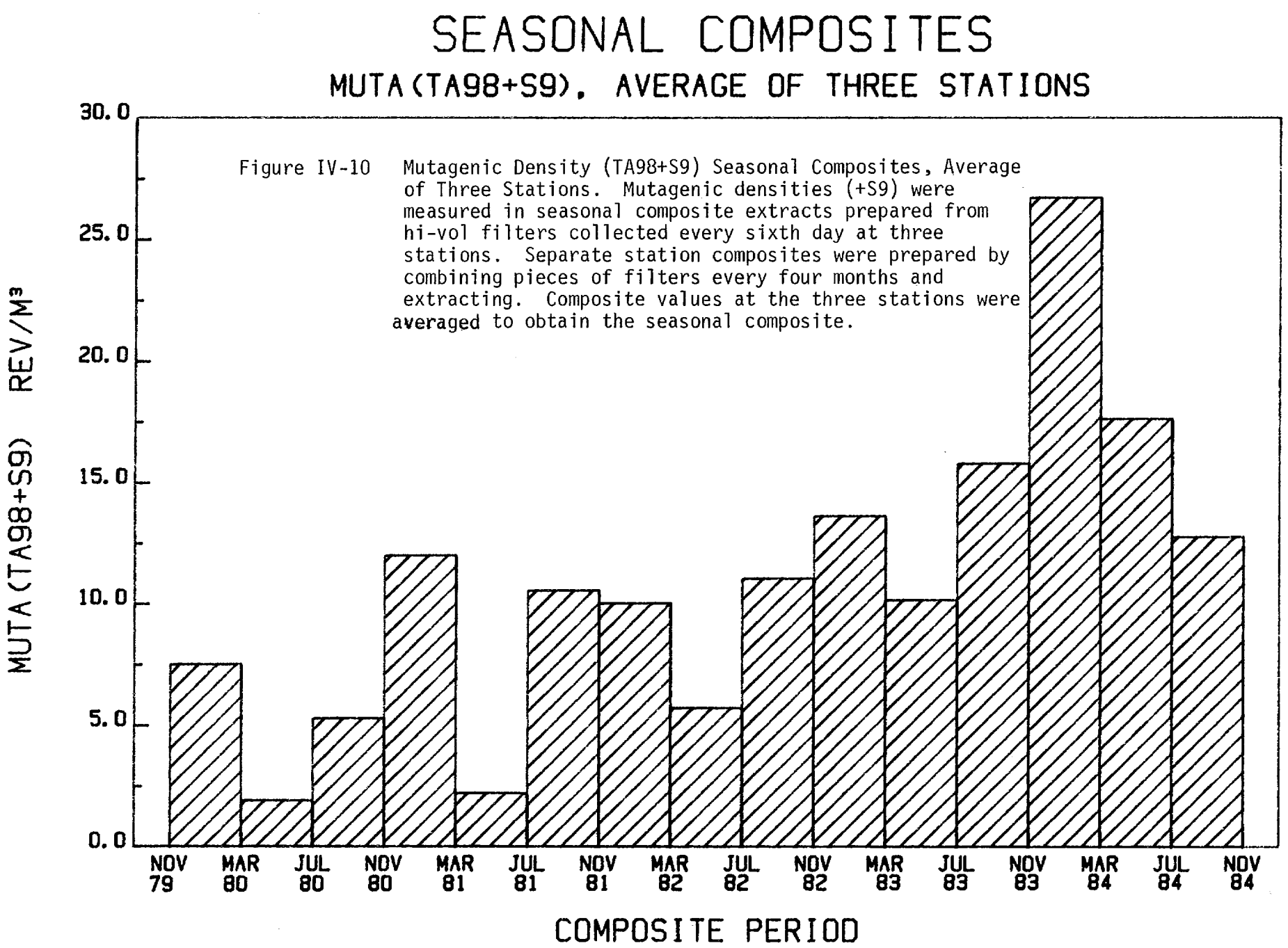
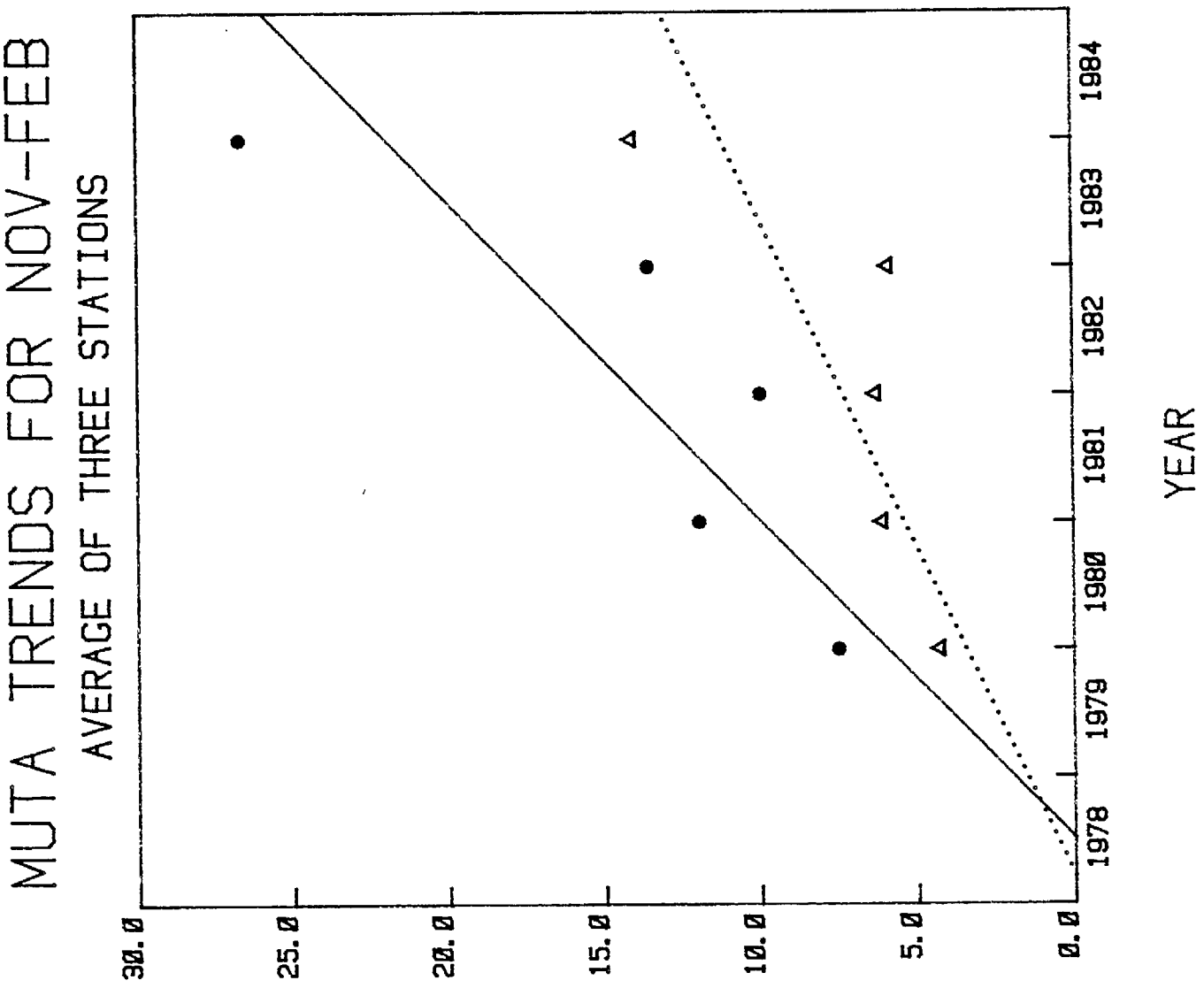


Figure IV-10 Mutagenic Density (TA98+S9) Seasonal Composites, Average of Three Stations. Mutagenic densities (+S9) were measured in seasonal composite extracts prepared from hi-vol filters collected every sixth day at three stations. Separate station composites were prepared by combining pieces of filters every four months and extracting. Composite values at the three stations were averaged to obtain the seasonal composite.

Figure IV-11

Mutagenic Density Trends for Nov.-Feb., Average of Three Stations. Trends in mutagenic density for the five winter seasons, 1979-1984 are compared by linear regression analysis. For TA98+S9, r=0.85 and b(slope)= 4.0 rev/yr. For TA98-S9, r=0.79 and b=1.9 rev/yr.



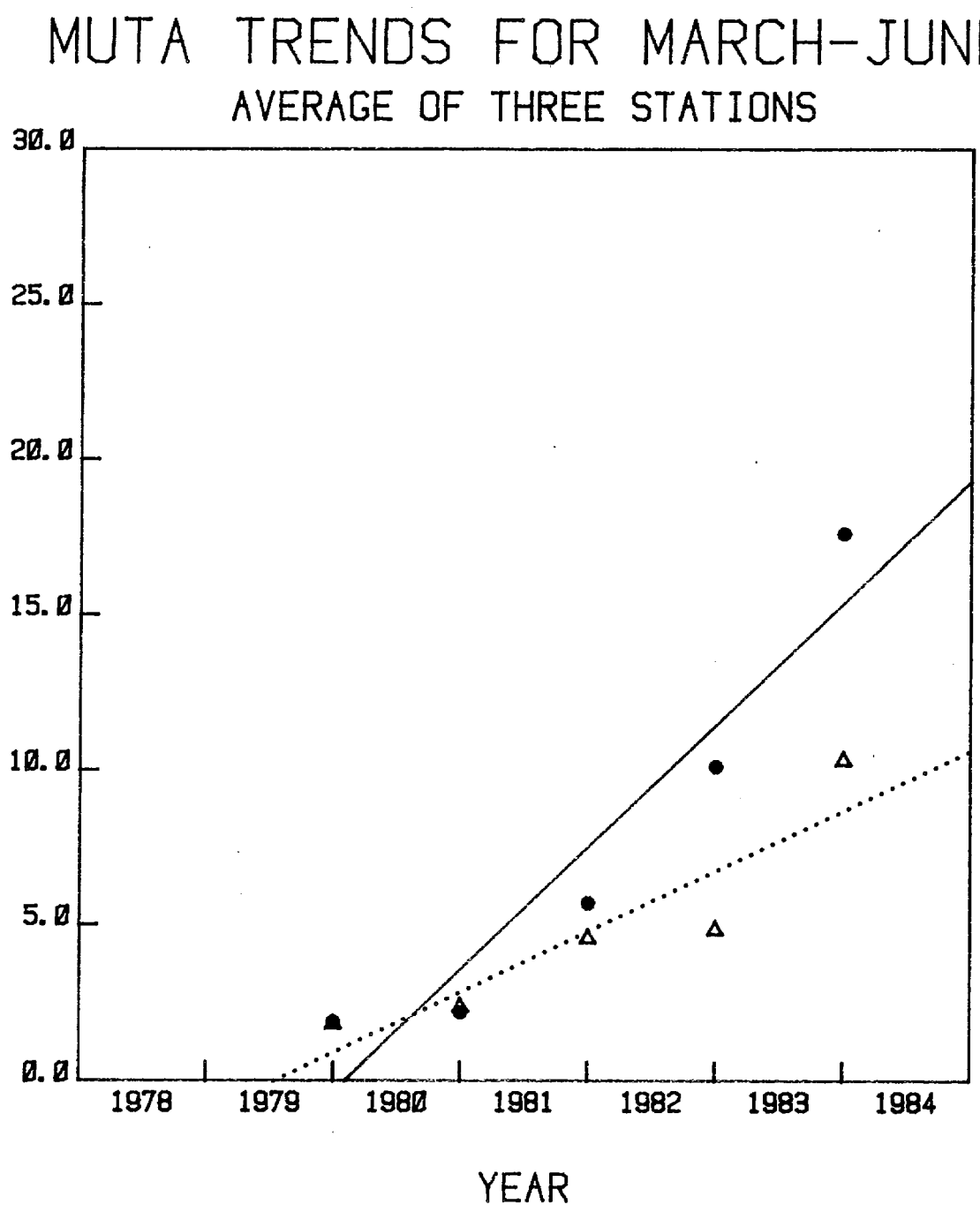


Figure IV-12

Mutagenic Density Trends for March-June, Average of Three Stations. Trends in mutagenic density for the five spring seasons, 1979-1984 are compared by linear regression analysis. For TA98+S9, r=0.95 and b(slope) = 3.9 rev/yr. For TA98-S9, r=0.91 and b = 1.9 rev/yr.

MUTA(+S9)
SYMBOL=●
LINETYPE=————
y=a+b*x
n=5
a=-7781.7600 s_a=1492.4784
b=3.9300 s_b=0.7530
$s_{y.x}$=2.3812 r=0.9491

MUTA(-S9)
SYMBOL=△
LINETYPE=.....
y=a+b*x
n=5
a=-3842.2800 s_a=981.2984
b=1.9410 s_b=0.4951
$s_{y.x}$=1.5657 r=0.9147

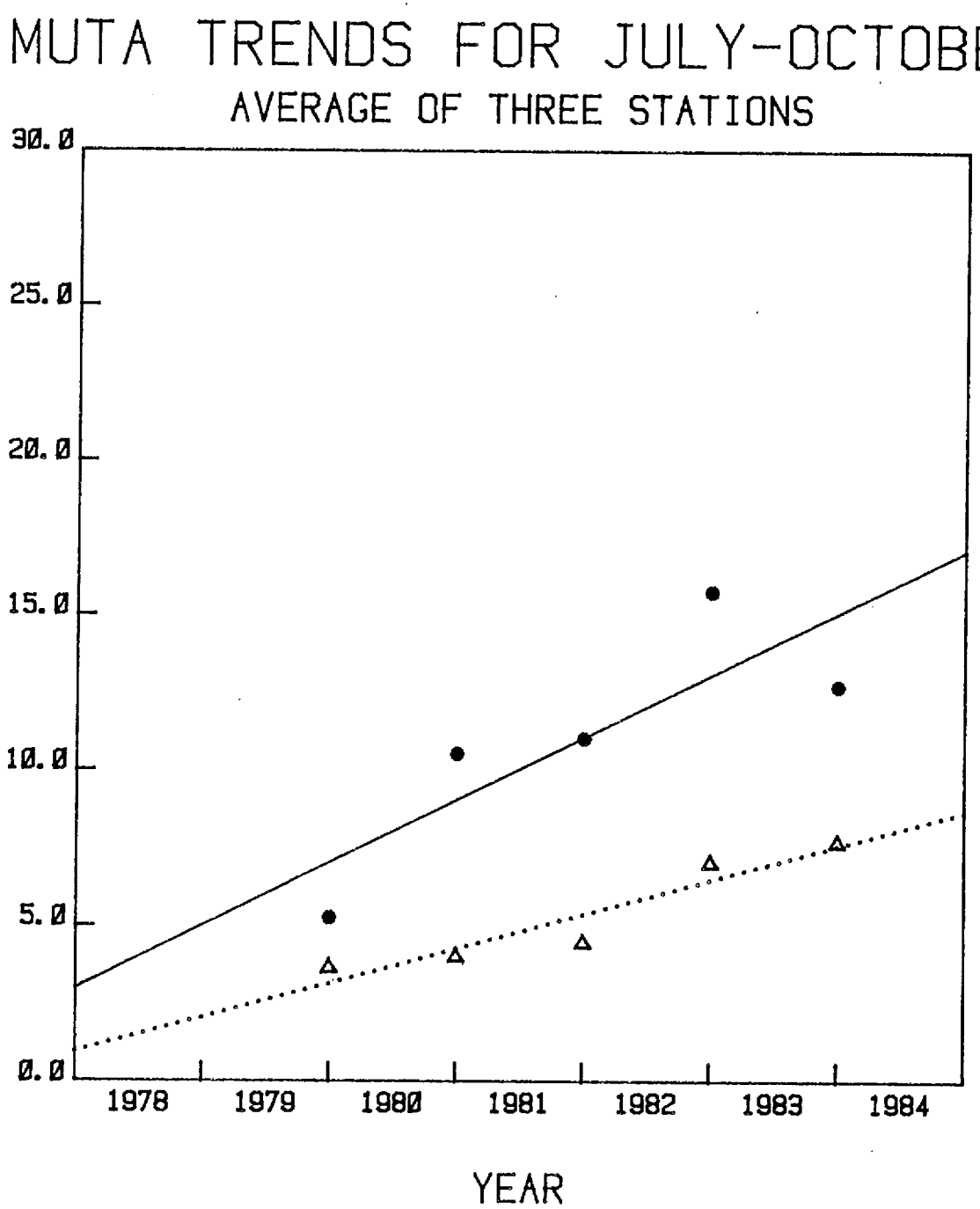


Figure IV-13

Mutagenic Density Trends for July-Oct., Average of Three Stations. Trends in mutagenic density for the five summer seasons, 1979-1984 are compared by linear regression analysis. For TA98+S9, r=0.83 and b(slope)= 2.0 rev/yr. For TA98-S9, r=0.95 and b=1.1 rev/yr.

MUTA (+S9)
SYMBOL=●
LINETYPE=————
y=a+b*x
n=5
a=-3984.6460 s_a=1541.0467
b=2.0160 s_b=0.7775
$s_{y.x}$=2.4587 r=0.8315

MUTA (-S9)
SYMBOL=△
LINETYPE=.....
y=a+b*x
n=5
a=-2186.7120 s_a=435.3248
b=1.1060 s_b=0.2196
$s_{y.x}$=0.6946 r=0.9456

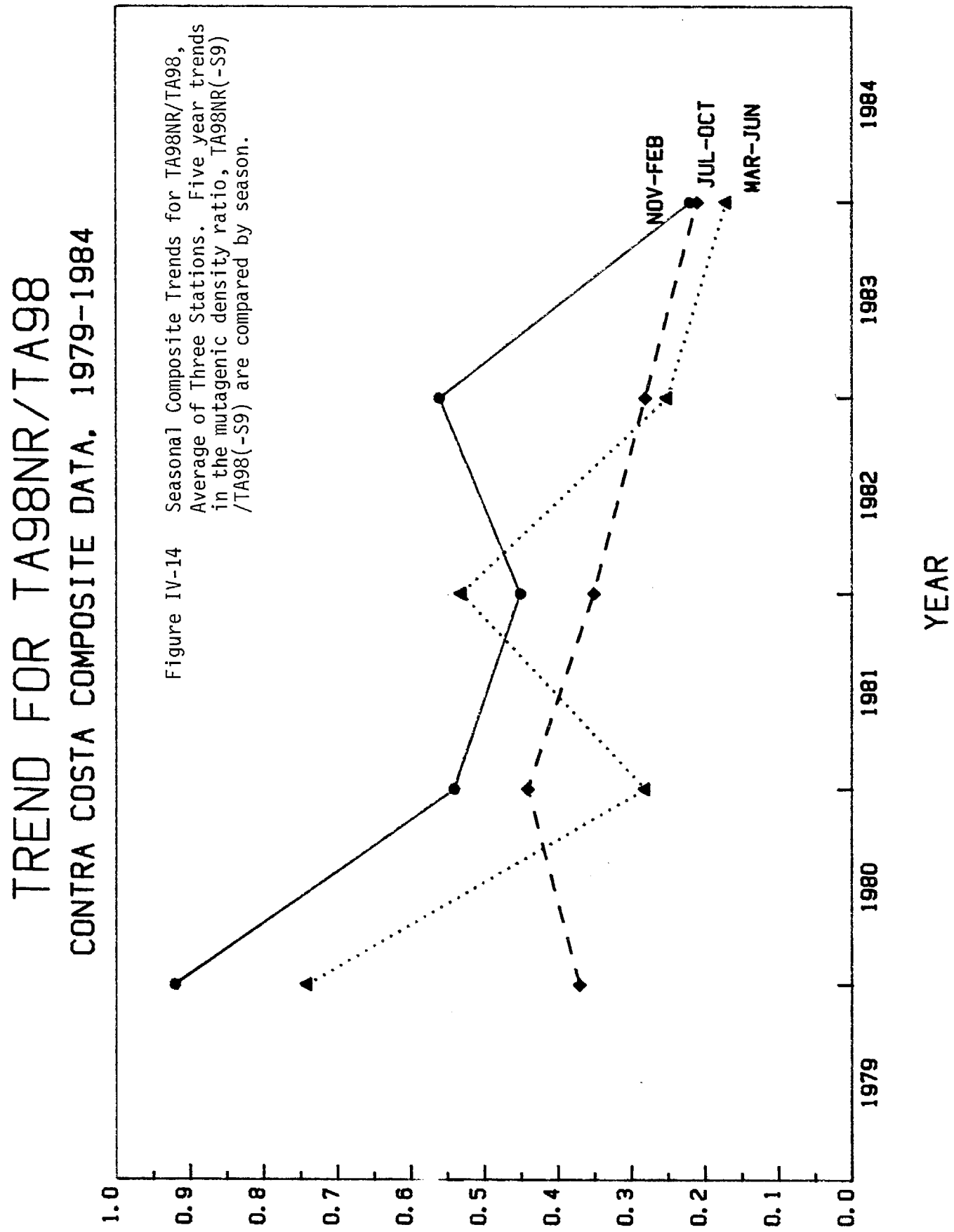


Figure IV-14 Seasonal Composite Trends for TA98NR/TA98, Average of Three Stations. Five year trends in the mutagenic density ratio, TA98NR(-S9) /TA98(-S9) are compared by season.

TABLE IV-3

COMPOSITE MUTAGENICITY TREND DATA

STRATIFIED BY LOCATION

Location	Mean Mutagenic Density in TA98+S9 by Year (3-Season Average) 1980 Mean	1980 S.D.	1981 Mean	1981 S.D.	1982 Mean	1982 S.D.	1983 Mean	1983 S.D.	1984 Mean	1984 S.D.
Pittsburg	3.6	(2.2)	5.2	(3.5)	10.1	(4.9)	11.4	(3.4)	19.5	(9.3)
Richmond	6.6	(5.0)	11.4	(8.4)	6.9	(4.1)	13.6	(2.0)	18.7	(8.2)
Concord	4.5	(3.1)	8.1	(6.0)	9.7	(6.4)	14.5	(3.4)	18.8	(14.0)
3-Station Average	4.9	(3.4)	8.2	(6.1)	8.9	(4.8)	13.1	(2.9)	19.0	(6.5)

S.D.= Standard Deviation

TABLE IV-4

COMPOSITE MUTAGENICITY DATA
STRATIFIED BY SEASON AND LOCATION

Season	Location	Mutagenic Density (TA98+S9) By Year 1980	1981	1982	1983	1984
Nov.-Feb.	Pittsburg	5.4	8.8	13.2	11.3	30.1
	Richmond	12.3	12.7	11.3	14.7	27.4
	Concord	4.8	14.4	5.5	14.8	22.6
Three Station Average		7.5	12.0	10.0	13.6	26.7
	S.D.	(4.2)	(2.9)	(4.0)	(2.0)	(3.8)
Mar.-June	Pittsburg	1.1	1.8	4.5	8.1	16.1
	Richmond	3.4	2.4	6.0	11.3	17.5
	Concord	1.2	2.4	6.6	11.0	19.2
Three Station Average		1.9	2.2	5.7	10.1	17.6
	S.D.	(1.3)	(0.3)	(1.1)	(1.8)	(1.6)
July-Oct.	Pittsburg	4.4	5.1	12.7	14.8	12.4
	Richmond	4.0	19.1	3.3	14.7	11.1
	Concord	7.4	7.4	17.1	17.8	14.7
Three Station Average		5.3	10.5	11.0	15.8	12.7
	S.D.	(1.9)	(7.5)	(7.0)	(1.8)	(1.8)

S.D. = Standard Deviation

TABLE IV-5

LINEAR REGRESSION ANALYSIS OF COMPOSITE MUTAGENICITY DATA (MUTAGENIC DENSITY IN TA98 + S9) YEAR VERSUS LOCATION AND SEASON

TA98 + S9 Versus Location	R^2	Slope (rev/yr)	F	Probability
. Pittsburg	0.92	3.8	36	0.009
. Richmond	0.69	2.6	7	0.08
. Concord	0.98	3.5	134	0.001
TA98 + S9 Versus Season				
. Nov.-Feb.	0.72	4.0	8	0.07
. March-June	0.90	3.9	27	0.01
. July-Oct.	0.69	2.0	7	0.08
Three Station (and Season) Average	0.93	3.3	41	0.008

on an annual basis. For NO_3^-, a statistically significant decrease also occurred, but only during the Nov.-Feb. season and on an annual basis. No other statistically significant changes were observed.

In contrast to the downward trends in some standard aerosol pollutants (Pb, NO_3^-) and the relative constancy of TSP, $SO_4^=$ and PAH (on an annual basis), mutagenic density exhibited an increasing trend over time (Figures IV-9-10). Concentrations of both direct-acting (-S9) and indirect-acting (+S9) mutagens increased over the study period, especially during the two seasons November 1983-June 1984. For example, during the five winter seasons, -S9 values increased from 4 to 14-rev/m^3 and +S9 values from 7.5 to 27 rev/m^3 (cf Figure IV-11). Similar trends in mutagenicity were observed during the spring (Figure IV-12) and summer (Figure IV-13).

The trend in mutagenicity can be analyzed in more detail by stratifying the composite data by location and season. Table IV-3 lists the (3-season average) mutagenic density (+S9) by location for the different years of the study. Qualitatively it is clear the increase in mutagenicity occurred at all three Contra Costa locations. Table IV-4 lists the mutagenic density (+S9) at each location by season for each year of the study. A nine-fold increase (from 2 to 18 revertants/m^3) occurred during the spring season, a 2-3 fold increase (from 5 to 13 revertants/m^3) occurred during the summer and a 3-4 fold increase (from 7.5 to 27 rev/m^3) in the winter, as noted above.

To provide further comparisons, linear regression analysis was carried out relating mutagenic density (+S9) with time both by station and by season. The results of linear regression analysis are listed in Table IV-5. The highest correlation between mutagenicity and time was at Concord (R^2 = 0.98) and the lowest at Richmond (R^2 = 0.70). Thus the trend is most uniform at Concord, a non-industrial location, and least uniform at Richmond, an industrial location most subject to marine influences. Concerning the seasonal time trends, the highest correlation occurred in the spring (R^2 = 0.90), when meteorlogical conditions are most uniform, and the lowest in the summer (R^2 = 0.69) and winter (R^2 = 0.72) when meteorological conditions are more variable.

Increasing mutagenic density may reflect larger contributions from NO_2-PAH. The possibility of an increasing impact over time of NO_2-PAH is suggested by a decreasing trend in the ratio of TA98NR/TA98 (Figure IV-14). This decrease suggests that NO_2-PAH are becoming more prominent contributors to the observed mutagenic density. Combustion related emissions are well known primary sources of nitroarenes, which may also be produced by secondary atmospheric reactions. The increase in mutagenic density may also be due in part to lower rainfall in the Bay Area during the first half of 1984. However it is not obvious how this could lead specifically to higher pollution levels of mutagenic aerosols and not other aerosol pollutants.

Regarding the trends in mutagenic density described above, some statements as to the consistency and quality control of filters, sample handling procedures, storage and mutagenic testing controls should be made. The first issue confounding the trend analysis concerns the filters used to collect the air particulate matter. Composites for Ames testing were prepared from particles collected on glass fiber filters used during routine monitoring by the Bay Area Air Quality Management District. The filters were purchased under EPA specification. Of possible relevance to the trend analysis is the fact that the filters actually used until December 1982 were Schleicher and Schwell #1-HV (S&S) while since January 1983, Whatman EPM 2000 hi-vol filters have been used. These two filters have large variations in alkalinity (49), which could amplify the artifact problem. As described earlier, gas phase HNO_3 can bind to alkaline sites on glass fiber and bound HNO_3 may catalyze chemical transformations of PAH to produce highly mutagenic nitroaromatic compounds during sampling collection. The available alkalinities varied by about a factor of two from 73 μ equiv/g for Whatman to 143 μ equiv/g for S&S filters (49). Fluctuations of this magnitude make attempts at trend analysis difficult. Nevertheless it should be noted that the expected impact of changing from higher pH S&S to lower pH Whatman filters is to <u>decrease</u> the potential for HNO_3-binding.

Following collections of filters by BAAQMD staff, the filters were transported to AIHL. Because of logistical and resource limitations, the time interval

between filter collection and delivery to the lab was typically 3-4 weeks, during which time the filters were held at room temperature. Once in the lab, within several days, pieces of filters for compositing were cut out and stored at -10°C in glassine envelopes wrapped in aluminum foil inside of zip-lock plastic bags. The time of cold storage of composite filters in this manner varied from several months to more than two years. No appropriate data for investigating the relationship between storage time and mutagenicity are available. Also, replicate analysis of filters from the same composite was not performed so the variability in the extraction and mutagenic assay of composites could not be assessed. However, an estimate of the experiment-to-experiment variability in the Ames assay itself can be obtained by comparing the variations in responses of positive control mutagens which were tested in parallel with the composites. The three positive controls used and their respective coefficients of variation over the study period were 2-aminofluorene, 28%; 2-nitrofluorene; 30%, and 4-nitro-quinoline-N-oxide, 30%. Based on these quality control data, we cannot rule out the possibility that methodological factors may explain the positive trend in mutagenic density.

Although detailed analysis of weather patterns over the study period is beyond the scope of this report, the following observations may provide some insight into the origins of the apparent increase in mutagenic density (Sandberg, J. personal communication). The use of weather factors to adjust trend studies has proved useful with ozone and of some value with carbon monoxide, but of limited value for particulate matter. The 24-hour basis of particulate measurements and the strong diurnal patterns (including wind direction reversals) typically observed in a 24-hour period in our complex terrain have made it difficult to isolate the weather factors most relevant for TSP on different types of days over the course of a year or series of years. However, the weather factors for ozone may be relevant for the photochemically related nitrate compounds (and nitroarenes). 1982 was a cool clean year, and 1983 and 1984 were very warm years with weaker than normal sea-breeze penetration related to the global El Nino event. Consequently, days over the Federal ozone standard did increase by a factor of four-from 5 in 1982 to 21 in 1983 and 22 in 1984. The ozone season is an extended summer event, but 1984 was particularly noteworthy for

its early ozone season, with mid-summer weather conditions observed in mid-April and in May. These months are classed in our analytic scheme with spring, which is normally cool, windy and clean. Also, the January and February weather factors for 1984 were atypically warm and dry.

Finally, we speculate that the actual changes in diesel emissions (50) which took place over the study period in Contra Costa County, especially in the vicinity of the sampling sites, probably did not account for a major proportion of the increase in mutagenic density. Detailed inventories of diesel emissions in the vicinity of the Contra Costa County sampling stations are being updated and prepared. The overall District diesel emissions do not rise sharply over the sampling period, but the expansion of the bus system in Contra Costa is being analyzed by BAAQMD staff for local impact.

D. Conclusions

The following conclusions may be drawn from the results of composite filter sampling carried out between November 1979-October 1984.

1. Seasonal comparisons indicate that higher values of mutagenic density, Pb, NO_3^-, and especially PAH were consistently observed in the winter seasons (November-February).

2. Decreasing (annual) trends in concentrations of Pb and NO_3^-, were also measured.

3. An increasing trend in the mutagenic density of Contra Costa aerosols was observed. The mutagenic density (rev/m^3) of Contra Costa community aerosols is three to four times higher in 1984 than it was in 1979. Further monitoring is needed to determine the persistence of this trend. Changes of this magnitude in pollution concentrations frequently can be explained by changes in wind direction and/or velocity. This is particularly true with small sample sizes. Perhaps this is also true for levels of mutagenicity.

In conclusion, we emphasize that in evaluating trends in air quality, analysts make one or both of two common assumptions:

a. Pollutant emissions are constant, hence the variations in pollutant concentrations are the result of some aspect of meteorological conditions.

b. Meteorological conditions while not constant, are effectively smoothed out when analyzing long term (i.e., several years) of data.

Since neither these assumptions is strictly valid, it is virtually impossible to establish true trends in pollutant concentrations, or its corollary, the effectiveness of control strategies, unless the function relationship between concentrations and meteorology has been determined and this we have not done. Only then will it be possible to utilize historical data for the determination of the true effectiveness of control strategies.

REFERENCES

1. Tokiwa H, Takeyoshi H, Morita K, Takahashi K, Saruta N, Ohnishi Y. (1976): Detection of mutagenic activity in urban air pollutants, Mutation Res., 38, 351-359

2. Talcott R, Wei E. (1977): Airborne mutagens bioassayed in Salmonella typhimurium, J. Nat. Cancer Inst., 58, 449-451.

3. Pitts J, Grosjean D, Mischke T, Simmon V, Poole D. (1977): Mutagencic activity of airborne particulate organic pollutants, Toxicology Letters, 1, 65-70.

4. Møller M and Alfheim I. (1980): Mutagencity and PAH-analysis of airborne particulate matter, Atmos. Environ., 14, 83-88.

5. Chrisp CE, Fisher GL. (1980): Mutagenicity of airborne particles. Mutation Res., 76:143-164.

6. Wesolowski J, Flessel P, Twiss S, Cheng J, Chan R, Garcia L, Ondo J, Fong A and Lum S. (1981): The chemical and biochemcial characterization of particulate matter as part of an epidemiological cancer study, J. Aerosol Sci., 12, 208-212.

7. Council on Environmental Quality (CEQ) (1980): Eleventh annual report of the Council on Environmental Quality. Washington, D.C.: U.S. Government Printing Office.

8. State of California Air Resources Board: A California Ambient Air Quality Standard for Particulate Matter (PM_{10}), Appendix 4, December 1982.

9. National Academy of Science (1972): Particulate polycyclic organic matter. Committee of biological effects of atmospheric pollutants, Washington D.C.

10. Gordon R, Bryan R, Rhim J, Demoise C, Wolford R, Freeman A, Heubner R. (1973): Transformation of rat and mouse embryo cells by a new class of carcinogenic compounds isolated from particles in city air. Int. J. Cancer., 12:233-232.

11. Pitts J: Formation and fate of gaseous and particulate mutagens and carcinogens in real and simulated atmospheres (1983): Environ. Health Perspec., 47:115-140.

12. Ames B, McCann J, Yamasaki E. (1975): Methods for detecting carcinogens and mutagens with the Salmonella/mammalian-microsome mutagenicity test, Mutation Res., 31, 347-364.

13. Pitts J, VanCauwenberge K, Grosjean D, Schmid J, Fitz D, Belser W, Knudson S, Hynds P: Atmospheric reactions of polycyclic aromatic hydrocarbons: Facile formation of mutagenic nitro derivatives (1978): Science, 202:515-519.

14. Schuetzle D, Perez J: Factors incluencing the emissions of nitrated-polynuclear aromatic hydrocarbons (Nitro-PAH) from diesel engines (1983): JAPCA, 33:751-755.

15. Wang Y, Lee M-S, King C, Warner P (1980): Evidence for nitro aromatics as direct-acting mutagens of airborne particulates Chemosphere, 9:83-87.

16. Siak J, Chan T, Gibson T, Wolf G. (1984): Contribution to bacterial mutagenicity from nitro-PAH compounds in ambient aerosols, paper 84-1.7 presented at the 77th Annual Meeting, Air Pollution Control Association, San Francisco, June 1984.

17. Pitts JN, Jr., Lokensgard DM, Fitz DR. (1982b): Chemical nature of particulate atmospheric mutagens in California's south coast air basin. Final Report: California Air Resources Board, Contract No. AO-139-32.

18. Flessel P, Guirguis G, Cheng J, Chang K, Hahn E, Chan R, Ondo J, Fenske R, Twiss S, Vance W, Wesolowski J, Kado N. (1984): Monitoring of Mutagens and Carcinogens in Community Air. Final Report: California Air Resources Board, Contract No. A1-029-32.

19. Kado NY, Langley D, Eisenstadt E. (1983): A simple modification of the Salmonella liquid incubation assay: increased sensitivity for detecting mutagens in human urine. Mutation Res., 121:25-32.

20. Gorse R, Riley F, Ferris F, Pero A, Skerves L. (1983): 1Nitropyrene concentrations and bacterial mutagenicity in on-road vehicle particulate emissions. Environ. Sci Technol., 17:198-202.

21. Gibson T. (1982): Nitro derivatives of polynuclear aromatic hydrocarbons in airborne and source particulate matter. Atmos. Environ. 16:2037-2040.

22. Sweetman J, Harger W, Fitz D, Paur HR, Winer A, Pitts J. (1984): Diurnal mutagenicity of airborne particulate organic matter adjacent to a heavily traveled West Los Angeles freeway, paper 84-16.5 presented at the 77th Annual Meeting, Air Pollution Control Association, San Francisco, June 1984.

23. Pitts J, Winer A, Sweetman J, et al. (1984): Particulate and Gas Phase Mutagens in Ambient and Simulated Atmospheres. Final Report: California Air Resources Board, Contract No. A3-049-32.

24. Shepson P, Kleindierst T, Edney E, Namie G, Pittman J, Cupitt L, Claxton L. (1985): The Mutagenic Activity of Irradiated Toluene/NOx/H_2O/Air Mixtures. Environ. Sci. Tecnol., 19:249-255.

25. Albrechcinski T, Michalovic J, Gibson T. (1984): Atmospheric reactions of polynuclear aromatic compounds as measured in a smog chamber. In Polynuclear Aromatic Hydrocarbons, edited by M. Cooke and A. Dennis, Battelle (in press).

26. Siak J, Chan T, Gibson T, Wolff G. (1985): Contribution to Bacterial Mutagenicity from Nitro-PAH Compounds in Ambient Aerosols. Atmos. Environ. 19:369-376.

27. Appel B, Tokiwa Y, Haik M, Kothny E. (1984): Artifact Particulate Sulfate and Nitrate Formation on Filter Media. Atmos. Environ. 18 409-416.

28. Bay Area Air Quality Management District Air Quality Handbook 1983-84. (1984) Bay Area Air Quality Management District, San Francisco, CA.

29. Pitts JN, Jr., Harger W, Lokensgard DM, Fitz DR, Scorziell GM, Mejia V. (1982a): Diurnal variations in the mutagenicity of airborne particulate organic matter in California's south coast air basin. Mutation Res., 104:35-41.

30. Grosjean D. (1983): Polycyclic aromatic hydrocarbons in Los Angeles air from samples collected on teflon, glass and quart filters. Atmospheric Environment, 17:2565-2573.

31. US EPA (1981): Quality Assurance Handbook for Air Pollution Measurement Systems. Vol. II Ambient Air Specific Methods. Revision No. 3, EPA-600/4-77-027a.

32. Loo BW, Adachi RS, Cork CP, Goulding FS, Jaklevic JM, Landis DA, Searles WL. (1979): A second generation dichotomous sampler for larger-scale monitoring of airborne particulate matter. LBL-8725 Presented at the 86th annual meeting of the American Institute of Chemical Engineers, Houston, Texas.

33. Flessel P, Wesolowski J, Twiss S, Cheng J, Ondo J, Monto N, Chan R. (1982): The integration of the Ames bioassay and chemical analyses in an epidemiological cancer incidence study, In: Second Symposium on Application of Short-term Bioassays in the Fractionation and Analysis of Complex Environmental Mixtures, (Waters M, ed.) New York, Plenum Press, pp. 61-84.

34. California Department of Health Services (1973): Determination of particulate lead. Method 41. Air and Industrial Hygiene Laboratory, Berkeley, CA.

35. McCann J, Springarn NE, Kobori J, Ames BN (1975): Detection of carcinogens as mutagens: bacterial tester strains with R factor plasmids. Proc. Natl. Acad. Sci. (USA), 72:979-983.

36. Rosenkranz HS, Speck WT. (1976): Activation of nitrofurantoin to a mutagen by rat liver nitroreductase. Biochem. Pharmacol., 25:1555-1556.

37. Lowry OH, Rosenbrough JN, Fan A, Randall RJ. (1951): Protein measurement with folin phenol reagent. J. Biol. Chem., 193:265-275.

38. Rosenkranz HS, Mermelstein R. (1983): Mutagenicity and genotoxicity of nitroarenes. All nitro-containing chemicals were not created equal. Mutation Res., 114:217-267.

39. Flessel CP, Guirguis GN, Cheng JC, Chang K, Hahn ES, Twiss S, Wesolowski JJ. (1985): Sources of mutagens in Contra Costa County community aerosols during pollution episodes: diurnal variations and relations to source emissions tracers. Environ. Internatl., (in press).

40. Talcott R, Harger W. (1980): Airborne mutagens extracted from particles of respirable size. Mutation Res., 79:177-180.

41. Sorenson WG, Whong W, Simpson JP, Hearl FJ, Ong T. (1982): Studies of the mutagenic response of Salmonella typhimurium TA98 to size-fractionated air particles: comparison of the fluctuation and plate incorporation tests. Environ. Mut., 4:531-541.

42. Giaque R, Goulding F, Jaklevic J, Pehl R. (1972): Trace element analysis with semiconductor detector x-ray spectrometers, LBL report-647 Technical Information Division, Lawrence Berkeley Laboratory.

43. Statistical Analysis System, User's Guide (1979): Helwig J. and Council, K. eds., SAS Institute Inc., Box 8000, Cary, North Carolina 27511.

44. Sexton K, Liu K, Hayward S, Spengler J. (1985): Characterization and source Apportionment of Wintertime Aerosol in a Wood-Burning Community. Atmosph. Environ. (in press).

45. Fitz D, Lokensgard D, Doyle G (1984): Investigation of Filtration Artifacts When Sampling Ambient Particulate Matter for Mutagen Assay. Atmosph. Environ., 18:205-213.

46. Appel, B, Wau S, Wesolowski J. (1976): "The Chemistry, Dispersion and Transport of Air Pollutants emitted from Fossil Fuel Power Plants in California," Final Report California Air Resources Board Research Contract No. ARB 3-948.

47. Rosenkranz E, McCoy E, Mermelstein R, Rosenkranz H. (1982): Evidence for Existence of Distinct Nitroreductases in Salmonella typhimurium: Roles in Mutagenesis, Carcinogenesis 3: 121-123.

48. Takeda N, Teranishi K, Hamada K. (1984): Mutagenicity of air pollutants collected at industrial, urban-residential and rural areas, Bull. Environ. Contamin. Toxicol., 32, 688-692.

49. Witz S, Smith M, Moore A (1983) = Comparative Performance of Glass Fiber Hi-Vol Filters. J. Air Poll. Control Assn. 33:988-991.

50. Wei E, Wang Y, Rappaport S.: Diesel emissions and the Ames test: A Commentary (1980): J. Air Pollut. Control Assoc., 30:267-271.

APPENDICES

APPENDIX I: Wind Speed and Direction at the Mountain View Sewage Treatment Plant, Martinez During Six Sampling Episodes (1982-1984).

APPENDIX II: San Francisco Bay Area Weather Factors During Six Sampling Episodes (1982-1984).

APPENDIX III: Complete Correlation Matrices for Combined Episodes, Daytime and Nighttime Samples, and the Four Stations.

APPENDIX IV: Complete Data Set for Contra Costa Seasonal Composites, Nov. 1979-Oct. 1984.

APPENDIX V: Linear Regression Slopes of Composite Aerosol Pollutant Data, 1979-1984. Year versus Season and Annual Average.

APPENDIX I:

WINDSPEED and DIRECTION at the
MOUNTAIN VIEW SEWAGE TREATMENT PLANT, MARTINEZ
DURING SIX SAMPLING EPISODES (1982 - 1984)

DATE	8/23/82			8/24/82	
PST	DRCTN	SPEED(mph)	PST	DRCTN	SPEED(mph)
			0100	255	9
			0200	240	7
0300	285	12	0300	240	8
0400	285	11	0400	240	8
0500	285	10	0500	240	7
0600	270	8	0600	240	8
0700	270	7	0700	240	7
0800	285	10	0800	255	7
0900	285	12	0900	270	11
1000	285	14	1000	270	13
1100	285	12	1100	270	14
1200	285	12	1200	285	13
1300	300	12	1300	285	13
1400	270	12	1400	285	12
1500	270	12	1500	270	11
1600	270	12	1600	255	10
1700	270	10	1700	255	9
1800	270	9	1800	270	10
1900	255	8	1900	270	9
2000	255	7	2000	240	7
2100	285	6	2100	210	3
2200	285	8	2200	270	6
2300	270	9	2300	240	2
2400	255	9	2400	60	1

APPENDIX I: (continued)

DATE 10/12/82			10/13/82			10/14/82		
PST	DRCTN	SPEED	PST	DRCTN	SPEED	PST	DRCTN	SPEED
			0100	195	2	0100	225	2
			0200	195	2	0200	225	2
			0300	240	3	0300	270	4
			0400	255	3	0400	270	4
			0500	240	3	0500	285	4
			0600	VRBL	1	0600	285	6
			0700	VRBL	1	0700	285	8
			0800	VRBL	1	0800	285	9
			0900	VRBL	1	0900	285	10
			1000	60	5	1000	285	10
			1100	45	6	1100	285	10
			1200	30	4	1200	285	10
			1300	30	6	1300	285	9
			1400	30	8			
1500	30	10	1500	45	5			
1600	45	8	1600	45	3			
1700	45	6	1700	345	2			
1800	60	2	1800	255	1			
1900	VRBL	1	1900	225	3			
2000	210	1	2000	270	3			
2100	VRBL	1	2100	270	6			
2200	VRBL	1	2200	285	3			
2300	210	1	2300	255	3			
2400	VRBL	1	2400	240	1			

APPENDIX I: (continued)

DATE	5/17/83		5/18/83			5/19/83		
PST	DRCTN	SPEED	PST	DRCTN	SPEED	PST	DRCTN	SPEED
			0100	255	4	0100	VRBL	1
			0200	255	4	0200	210	1
			0300	255	3	0300	150	1
			0400	240	2	0400	195	2
			0500	195	1	0500	VRBL	1
			0600	VRBL	1	0600	210	2
			0700	240	2	0700	VRBL	1
			0800	240	3	0800	VRBL	1
			0900	270	5	0900	VRBL	1
			1000	300	5	1000	VRBL	2
			1100	300	4	1100	030	8
			1200	315	5	1200	030	9
			1300	300	3	1300	030	10
			1400	300	5	1400	030	10
			1500	300	5	1500	030	8
1600	360	5	1600	300	6	1600	030	6
1700	300	7	1700	300	6	1700	030	6
1800	285	8	1800	285	4	1800	330	2
1900	285	7	1900	285	5	1900	300	5
2000	270	3	2000	285	6	2000	285	6
2100	VRBL	1	2100	270	6	2100	285	6
2200	VRBL	1	2200	270	5	2200	225	3
2300	VRBL	1	2300	270	3	2300	210	1
2400	255	4	2400	VRBL	1	2400	VRBL	1

APPENDIX I: (continued)

DATE	9/12/83		9/13/83			9/14/83		
PST	DRCTN	SPEED	PST	DRCTN	SPEED	PST	DRCTN	SPEED
			0100	240	2	0100	270	5
			0200	VRBL	1	0200	270	6
			0300	VRBL	<1	0300	270	6
			0400	255	1	0400	270	5
			0500	270	2	0500	270	7
			0600	VRBL	1	0600	270	7
			0700	VRBL	<1	0700	270	7
			0800	300	3	0800	270	7
			0900	285	7			
			1000	285	8			
			1100	300	9			
			1200	300	9			
			1300	300	10			
			1400	300	10			
			1500	285	10			
			1600	285	9			
1700	360	4	1700	270	9			
1800	360	4	1800	270	9			
1900	300	3	1900	285	8			
2000	VRBL	1	2000	270	8			
2100	300	2	2100	270	8			
2200	300	4	2200	285	4			
2300	285	4	2300	270	3			
2400	300	2	2400	270	7			

APPENDIX I: (continued)

DATE	10/4/83		10/5/83			10/6/83		
PST	DRCTN	SPEED	PST	DRCTN	SPEED	PST	DRCTN	SPEED
			0100	VRBL	1	0100	255	7
			0200	225	2	0200	240	8
			0300	150	2	0300	240	5
			0400	195	2	0400	255	5
			0500	255	1	0500	255	6
			0600	240	2	0600	210	2
			0700	210	1	0700	255	5
			0800	240	3	0800	270	7
			0900	300	4	0900	240	5
			1000	300	5	1000	315	5
			1100	270	9	1100	315	4
			1200	270	9	1200	315	3
			1300	240	10	1300	345	4
			1400	240	8	1400	360	3
			1500	240	7	1500	360	4
			1600	225	8	1600	345	3
1700	285	5	1700	225	9			
1800	270	2	1800	240	5			
1900	270	5	1900	225	8			
2000	270	6	2000	255	8			
2100	270	3	2100	255	4			
2200	VRBL	1	2200	270	7			
2300	MISSING		2300	270	7			
2400	MISSING		2400	255	7			

APPENDIX I: (continued)

DATE	1/4/84		1/5/84			1/6/84		
PST	DRCTN	SPEED	PST	DRCTN	SPEED	PST	DRCTN	SPEED
			0100	060	6	0100	045	7
			0200	060	7	0200	045	8
			0300	060	5	0300	045	8
			0400	060	6	0400	045	8
			0500	045	6	0500	045	8
			0600	045	6	0700	060	7
			0700	045	6	0800	045	6
			0800	045	6			
			0900	045	5			
			1000	045	6			
			1100	060	6			
			1200	045	7			
			1300	045	6			
			1400	060	7			
			1500	030	6			
			1600	015	5			
1700	015	4	1700	030	6			
1800	015	4	1800	030	5			
1900	015	4	1900	030	6			
2000	030	4	2000	030	5			
2100	030	5	2100	045	6			
2200	030	5	2200	045	7			
2300	030	6	2300	045	5			
2400	030	6	2400	045	6			

APPENDIX II: SAN FRANCISCO BAY AREA WEATHER FACTORS* DURING SIX SAMPLING EPISODES 1982-84.

BAY AREA WEATHER FACTORS
Include:

Mean wind speed in mph for Central (C) from SFO airport, for North from BAAQMD Vallejo (VA) station, for South from BAAQMD San Jose (SJ) station.

Mean max, temperature (deg. F) for C averaged from SFO and SF, for North from San Rafael (SR), for South from SJ.

Total insolation, in Langleys/day, as measured by Eppley pyranometer.

Ventilation from OAK radiosonde data on 1 to 5 scale of increasing intensity, with airflow direction at 1000 millibar level.

Stability factor is temp. (deg. F) at 2500 feet minus that at the surface, representing low-level inversion strength at Oakland OA, Condord CC and SJ. Vertical mixing decreases with algebraic value of stability factor.

*These data published monthly by the Bay Area Air Quality Management District, Technical Services Division 117 in "Contaminant and Weather Summary".

APPENDIX II:
SAN FRANCISCO BAY AREA WEATHER FACTORS DURING 1982-1984 EPISODES

Date	Mean wind Speed (mph)			Max. Temp. (F)			Insolation (LY/day)			Ventilation		Stability Factor		
	C	VA	SJ	C	SR	SJ	SF	SR	SJ	4am	4pm	OA	CC	SJ
August 1982	14.4	6.8	3.4	71	81	84	259	435	415	NW2	W2	25	20	17
M 23	14.4	6.8	3.4	71	81	84	259	435	415	NW2	W2	25	20	17
T 24	12.2	6.7	3.1	63	79	77	185	438	380	W2	W2	9	9	3
Oct 1982														
T 12	9.6	2.7	1.5	76	84	86	265	288	265	LVI		12	18	9
W 13	10.9	2.3	1.7	76	87	88	265	270	275		LVI			
Th 14	15.1	2.3	1.7	62	74	75	202	222	215	WI	NWI	12	15	9
May 1983														
T 17	7.8	3.2	3.1	82	88	86	460	516	500	NI	W2	15	13	6
W 18	12.1	2.9	3.4	76	87	90	433	492	485	SEI	NWI	14	15	8
TH 19	6.6	3.0	3.4	88	92	90	469	528	490	LVI	NES	10	8	2
September 1983														
M 12	7.5	2.8		98	101	99	359	396		LVI	NWI	17	22	12
T 13	15.0	4.3		78	91	89	369	444		WI	W2	10	19	4
W 14	10.9	2.4		84	92	93	372	420		NWI	NWI	20	21	13
Oct. 1983														
W 5	8.3	4.9	3.0	74	78	86	282	351	306	SI	W3	10	15	15
TH 6	9.4	2.8	2.2	78	80	82	274	336	227	NW2	W1	6	13	13
Jan 1984														
W 4	7.9	8.1	1.0	58	54	62	141	144	125	EI	NE2	8	11	9
TH 5	8.2	6.7	1.4	54	51	57	142	72	110	NEI	SE2	14	15	10
F 6	8.0	7.6	1.0	54	49	56	160	81	85	EI	NW2	19	20	14

APPENDIX III: COMPLETE CORRELATION MATRICES FOR COMBINED EPISODES, DAYTIME AND NIGHTTIME SAMPLES AND THE FOUR STATIONS.

VARIABLE	N	MEAN	STD DEV	SUM	MINIMUM	MAXIMUM
TA98P	72	19.84166665	12.01082550	1428.5999988	0.60000000	58.39999998
TA98M	72	9.14166666	5.67484565	658.1999995	0.40000000	26.79999995
BAP	72	0.20000000	0.24321786	14.4000000	0.10000000	1.60000000
COR	72	0.58055556	0.39881475	41.8000000	0.10000000	1.60000000
BO	72	0.75000000	0.85826553	54.0000000	0.10000000	5.10000000
PBF	72	239.69152778	177.68068817	17257.7900000	17.00000000	950.00000000
BRF	72	44.50416667	34.50425341	3204.3000000	6.00000000	184.00000000
KF	72	136.10416667	105.51221468	9799.5000000	31.00000000	491.00000000
ZNF	72	24.71250000	19.20486248	1779.3000000	3.00000000	123.00000000
FEF	72	125.33472222	101.30239095	9024.1000000	12.00000000	450.00000000
SIF	72	283.35000000	242.67248839	20401.2000000	24.00000000	1220.00000000
CLF	72	288.53888889	527.75181308	20774.8000000	12.00000000	3050.00000000
NIF	72	7.79722222	8.83981027	561.4000000	2.00000000	48.00000000
SF	72	1794.80000000	1319.22627505	129225.6000000	442.00000000	7640.00000000
NO3	71	7.59577465	4.67578676	539.2999998	3.00000000	24.50000000
CO	54	1.09999985	0.46702578	59.3999917	0.19999999	2.79999924
NO	63	1.79047593	1.46702605	112.7999833	0	6.39999962
NO2	69	2.59420248	1.27543350	178.9999712	0.09999996	6.29999924
O3	69	1.99565192	1.45535861	137.6999827	0	5.89999962
SO2	69	0.36811586	0.75078232	25.3999946	0	4.29999924

CORRELATIONS OF COMBINED EPISODE DATA, pp 1-5

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
TA98P	1.00000	0.94186	0.45133	0.54114	0.60456	0.27814	0.39989	0.26186	0.11718	0.08699	-0.01756	-0.26919	-0.09966
	0.0000	0.0001	0.0001	0.0001	0.0001	0.0180	0.0005	0.0263	0.3270	0.4675	0.8836	0.0222	0.4049
	72	72	72	72	72	72	72	72	72	72	72	72	72
TA98M	0.94186	1.00000	0.48788	0.43885	0.59108	0.24562	0.37749	0.21840	0.12304	0.12537	0.01425	-0.22215	-0.07457
	0.0001	0.0000	0.0001	0.0001	0.0001	0.0376	0.0011	0.0653	0.3032	0.2940	0.9054	0.0607	0.5336
	72	72	72	72	72	72	72	72	72	72	72	72	72
BAP	0.45133	0.48788	1.00000	0.37608	0.92099	0.07810	0.30205	0.15420	-0.00763	-0.07278	-0.06918	0.01398	-0.11687
	0.0001	0.0001	0.0000	0.0011	0.0001	0.5144	0.0099	0.1959	0.9493	0.5435	0.5636	0.9072	0.3282
	72	72	72	72	72	72	72	72	72	72	72	72	72
COR	0.54114	0.43885	0.37608	1.00000	0.55756	0.70548	0.73679	0.58534	0.26894	0.28486	0.13409	-0.14507	-0.20329
	0.0001	0.0001	0.0011	0.0000	0.0001	0.0001	0.0001	0.0001	0.0224	0.0153	0.2615	0.2240	0.0868
	72	72	72	72	72	72	72	72	72	72	72	72	72
BO	0.60456	0.59108	0.92099	0.55756	1.00000	0.19453	0.43866	0.25566	0.08036	-0.05102	-0.08937	-0.05174	-0.11664
	0.0001	0.0001	0.0001	0.0001	0.0000	0.1015	0.0001	0.0302	0.5022	0.6704	0.4553	0.6660	0.3292
	72	72	72	72	72	72	72	72	72	72	72	72	72
PBF	0.27814	0.24562	0.07810	0.70548	0.19453	1.00000	0.90353	0.83143	0.49237	0.73638	0.52051	-0.14795	0.18383
	0.0180	0.0376	0.5144	0.0001	0.1015	0.0000	0.0001	0.0001	0.0001	0.0001	0.0001	0.2149	0.1222
	72	72	72	72	72	72	72	72	72	72	72	72	72
BRF	0.39989	0.37749	0.30205	0.73679	0.43866	0.90353	1.00000	0.82557	0.42557	0.60402	0.40616	-0.01936	0.14256
	0.0005	0.0011	0.0099	0.0001	0.0001	0.0001	0.0000	0.0001	0.0002	0.0001	0.0004	0.8718	0.2322
	72	72	72	72	72	72	72	72	72	72	72	72	72
KF	0.26186	0.21840	0.15420	0.58534	0.25566	0.83143	0.82557	1.00000	0.43057	0.69375	0.56748	0.00848	0.20696
	0.0263	0.0653	0.1959	0.0001	0.0302	0.0001	0.0001	0.0000	0.0002	0.0001	0.0001	0.9437	0.0811
	72	72	72	72	72	72	72	72	72	72	72	72	72
ZNF	0.11718	0.12304	-0.00763	0.26894	0.08036	0.49237	0.42557	0.43057	1.00000	0.51986	0.30368	0.07131	0.30021
	0.3270	0.3032	0.9493	0.0224	0.5022	0.0001	0.0002	0.0002	0.0000	0.0001	0.0095	0.5516	0.0104
	72	72	72	72	72	72	72	72	72	72	72	72	72
FEF	0.08699	0.12537	-0.07278	0.28486	-0.05102	0.73638	0.60402	0.69375	0.51986	1.00000	0.87518	-0.13744	0.44345
	0.4675	0.2940	0.5435	0.0153	0.6704	0.0001	0.0001	0.0001	0.0001	0.0000	0.0001	0.2496	0.0001
	72	72	72	72	72	72	72	72	72	72	72	72	72
SIF	-0.01756	0.01425	-0.06918	0.13409	-0.08937	0.52051	0.40616	0.56748	0.30368	0.87518	1.00000	-0.10739	0.37160
	0.8836	0.9054	0.5636	0.2615	0.4553	0.0001	0.0004	0.0001	0.0095	0.0001	0.0000	0.3692	0.0013
	72	72	72	72	72	72	72	72	72	72	72	72	72
CLF	-0.26919	-0.22215	0.01398	-0.14507	-0.05174	-0.14795	-0.01936	0.00848	0.07131	-0.13744	-0.10739	1.00000	-0.08486
	0.0222	0.0607	0.9072	0.2240	0.6660	0.2149	0.8718	0.9437	0.5516	0.2496	0.3692	0.0000	0.4785
	72	72	72	72	72	72	72	72	72	72	72	72	72
NIF	-0.09966	-0.07457	-0.11687	-0.20329	-0.11664	0.18383	0.14256	0.20696	0.30021	0.44345	0.37160	-0.08486	1.00000
	0.4049	0.5336	0.3282	0.0868	0.3292	0.1222	0.2322	0.0811	0.0104	0.0001	0.0013	0.4785	0.0000
	72	72	72	72	72	72	72	72	72	72	72	72	72

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
SF	0.21728	0.23319	0.27653	0.03011	0.36289	0.03302	0.24073	0.14947	0.14618	-0.07158	-0.03584	-0.03273	0.21207
	0.0667	0.0487	0.0187	0.8017	0.0017	0.7830	0.0417	0.2101	0.2205	0.5502	0.7650	0.7849	0.0737
	72	72	72	72	72	72	72	72	72	72	72	72	72
NO3	0.49496	0.51066	0.32107	0.14935	0.47658	-0.03134	0.16413	0.16149	0.22443	-0.08395	-0.07644	-0.01536	0.04641
	0.0001	0.0001	0.0063	0.2138	0.0001	0.7953	0.1714	0.1785	0.0599	0.4864	0.5264	0.8989	0.7007
	71	71	71	71	71	71	71	71	71	71	71	71	71
CO	0.54527	0.51478	0.22569	0.71090	0.36294	0.63471	0.66256	0.49970	0.33739	0.40650	0.20033	-0.05903	0.25321
	0.0001	0.0001	0.1008	0.0001	0.0070	0.0001	0.0001	0.0001	0.0126	0.0023	0.1464	0.6716	0.0647
	54	54	54	54	54	54	54	54	54	54	54	54	54
NO	0.51037	0.44927	0.33604	0.49429	0.44249	0.48813	0.64595	0.37869	0.12802	0.27377	0.19516	-0.15416	0.14321
	0.0001	0.0002	0.0071	0.0001	0.0003	0.0001	0.0001	0.0022	0.3174	0.0299	0.1253	0.2277	0.2628
	63	63	63	63	63	63	63	63	63	63	63	63	63
NO2	0.32113	0.31262	0.10563	0.60602	0.17894	0.82420	0.69757	0.60710	0.43082	0.63826	0.47300	-0.12231	0.24964
	0.0071	0.0089	0.3877	0.0001	0.1413	0.0001	0.0001	0.0001	0.0002	0.0001	0.0001	0.3167	0.0386
	69	69	69	69	69	69	69	69	69	69	69	69	69
O3	-0.27144	-0.35349	-0.40103	-0.12963	-0.43105	-0.00401	-0.21271	0.04710	0.11161	0.18520	0.19501	-0.05802	0.07773
	0.0241	0.0029	0.0006	0.2884	0.0002	0.9739	0.0793	0.7007	0.3613	0.1276	0.1083	0.6358	0.5255
	69	69	69	69	69	69	69	69	69	69	69	69	69
SO2	0.04063	0.05412	-0.02478	-0.27496	-0.02399	-0.08152	0.06775	0.01647	0.04868	0.06690	0.12398	-0.03378	0.33455
	0.7403	0.6587	0.8398	0.0222	0.8449	0.5055	0.5802	0.8932	0.6912	0.5850	0.3101	0.7829	0.0050
	69	69	69	69	69	69	69	69	69	69	69	69	69

	SF	NO3	CO	NO	NO2	O3	SO2
TA98P	0.21728	0.49496	0.54527	0.51037	0.32113	-0.27144	0.04063
	0.0667	0.0001	0.0001	0.0001	0.0071	0.0241	0.7403
	72	71	54	63	69	69	69
TA98M	0.23319	0.51066	0.51478	0.44927	0.31262	-0.35349	0.05412
	0.0487	0.0001	0.0001	0.0002	0.0089	0.0029	0.6587
	72	71	54	63	69	69	69
BAP	0.27653	0.32107	0.22569	0.33604	0.10563	-0.40103	-0.02478
	0.0187	0.0063	0.1008	0.0071	0.3877	0.0006	0.8398
	72	71	54	63	69	69	69
COR	0.03011	0.14935	0.71090	0.49429	0.60602	-0.12963	-0.27496
	0.8017	0.2138	0.0001	0.0001	0.0001	0.2884	0.0222
	72	71	54	63	69	69	69
BO	0.36289	0.47658	0.36294	0.44249	0.17894	-0.43105	-0.02399
	0.0017	0.0001	0.0070	0.0003	0.1413	0.0002	0.8449
	72	71	54	63	69	69	69
PBF	0.03302	-0 03134	0.63471	0.48813	0.82420	-0.00401	-0.08152
	0.7830	0.7953	0.0001	0.0001	0.0001	0.9739	0.5055
	72	71	54	63	69	69	69

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	SF	NO3	CO	NO	NO2	O3	SO2
BRF	0.24073	0.16413	0.66256	0.64595	0.69757	-0.21271	0.06779
	0.0417	0.1714	0.0001	0.0001	0.0001	0.0793	0.5802
	72	71	54	63	69	69	69
KF	0.14947	0.16149	0.49970	0.37869	0.60710	0.04710	0.01647
	0.2101	0.1785	0.0001	0.0022	0.0001	0.7007	0.8932
	72	71	54	63	69	69	69
ZNF	0.14618	0.22443	0.33739	0.12802	0.43082	0.11161	0.04868
	0.2205	0.0599	0.0126	0.3174	0.0002	0.3613	0.6912
	72	71	54	63	69	69	69
FEF	-0.07158	-0.08395	0.40650	0.27377	0.63826	0.18520	0.06690
	0.5502	0.4864	0.0023	0.0299	0.0001	0.1276	0.5850
	72	71	54	63	69	69	69
SIF	-0.03584	-0.07644	0.20033	0.19516	0.47300	0.19501	0.12398
	0.7650	0.5264	0.1464	0.1253	0.0001	0.1083	0.3101
	72	71	54	63	69	69	69
CLF	-0.03273	-0.01536	-0.05903	-0.15416	-0.12231	-0.05802	-0.03378
	0.7849	0.8989	0.6716	0.2277	0.3167	0.6358	0.7829
	72	71	54	63	69	69	69
NIF	0.21207	0.04641	0.25321	0.14321	0.24964	0.07773	0.33455
	0.0737	0.7007	0.0647	0.2628	0.0386	0.5255	0.0050
	72	71	54	63	69	69	69
SF	1.00000	0.46209	0.09243	0.12183	-0.02183	-0.26197	0.30023
	0.0000	0.0001	0.5062	0.3415	0.8587	0.0297	0.0122
	72	71	54	63	69	69	69
NO3	0.46209	1.00000	0.22485	0.16421	-0.03172	-0.29535	0.38607
	0.0001	0.0000	0.1055	0.2022	0.7973	0.0145	0.0011
	71	71	53	62	68	68	68
CO	0.09243	0.22485	1.00000	0.67991	0.55757	-0.07620	0.12504
	0.5062	0.1055	0.0000	0.0001	0.0001	0.5839	0.3676
	54	53	54	54	54	54	54
NO	0.12183	0.16421	0.67991	1.00000	0.41523	-0.27091	0.37215
	0.3415	0.2022	0.0001	0.0000	0.0007	0.0317	0.0027
	63	62	54	63	63	63	63
NO2	-0.02183	-0.03172	0.55757	0.41523	1.00000	-0.08986	-0.02262
	0.8587	0.7973	0.0001	0.0007	0.0000	0.4628	0.8536
	69	68	54	63	69	69	69
O3	-0.26197	-0.29535	-0.07620	-0.27091	-0.08986	1.00000	-0.18398
	0.0297	0.0145	0.5839	0.0317	0.4628	0.0000	0.1302
	69	68	54	63	69	69	69

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	SF	NO3	CO	NO	NO2	O3	SO2
SO2	0.30023	0.38607	0.12504	0.37215	-0.02262	-0.18398	1.00000
	0.0122	0.0011	0.3676	0.0027	0.8536	0.1302	0.0000
	69	68	54	63	69	69	69

DN=D

VARIABLE	N	MEAN	STD DEV	SUM	MINIMUM	MAXIMUM
TA98P	28	22.02499999	13.65803589	616.69999959	4.40000000	58.39999998
TA98M	28	9.28214285	6.20836076	259.89999982	1.00000000	26.79999995
BAP	28	0.16785714	0.09449112	4.70000000	0.10000000	0.40000000
COR	28	0.70357143	0.41229452	19.69999999	0.10000000	1.60000000
BO	28	0.74285714	0.57634132	20.79999999	0.10000000	2.00000000
PBF	28	256.22607143	148.83820421	7174.33000000	49.00000000	638.00000000
BRF	28	46.05357143	29.94573465	1289.50000000	8.00000000	114.00000000
KF	28	143.58214286	123.68640170	4020.30000000	36.00000000	491.00000000
ZNF	28	31.75714286	22.74413931	889.20000000	9.00000000	123.00000000
FEF	28	135.96428571	95.12233143	3807.00000000	28.00000000	371.00000000
SIF	28	306.18571429	268.59415028	8573.20000000	24.00000000	1220.00000000
CLF	28	174.68928571	261.58089088	4891.30000000	19.00000000	1450.00000000
NIF	28	8.08928571	9.72973066	226.50000000	2.00000000	48.00000000
SF	28	1791.72500000	1285.52283891	50168.30000000	442.00000000	7640.00000000
NO3	27	7.98148148	3.53129275	215.49999996	3.20000000	15.80000000
CO	21	1.21428562	0.39406297	25.49999797	0.59999996	2.09999943
NO	24	2.04166641	1.45659409	48.99999392	0.50000000	6.39999962
NO2	27	2.58148094	1.05138633	69.69998550	1.19999981	5.29999924
O3	27	2.88888863	1.38933659	77.99999291	0	5.89999962
SO2	27	0.41851842	0.87179595	11.29999745	0	4.29999924

CORRELATIONS OF DAYTIME EPISODE DATA, pp 6-10

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
TA98P	1.00000 0.0000 28	0.94822 0.0001 28	0.52984 0.0037 28	0.50242 0.0064 28	0.64686 0.0002 28	0.19471 0.3208 28	0.37528 0.0491 28	0.24386 0.2111 28	-0.14260 0.4691 28	-0.11611 0.5563 28	-0.11611 0.5563 28	-0.19705 0.3149 28	-0.32613 0.0903 28
TA98M	0.94822 0.0001 28	1.00000 0.0000 28	0.57477 0.0014 28	0.37739 0.0477 28	0.59157 0.0009 28	0.06759 0.7326 28	0.29613 0.1260 28	0.09965 0.6139 28	-0.19311 0.3248 28	-0.18015 0.3590 28	-0.16029 0.4152 28	-0.12647 0.5213 28	-0.38751 0.0416 28
BAP	0.52984 0.0037 28	0.57477 0.0014 28	1.00000 0.0000 28	0.50692 0.0059 28	0.76753 0.0001 28	0.27670 0.1540 28	0.58768 0.0010 28	0.39484 0.0376 28	-0.18851 0.3367 28	0.04107 0.8356 28	0.14089 0.4745 28	-0.08588 0.6639 28	-0.19215 0.3273 28
COR	0.50242 0.0064 28	0.37739 0.0477 28	0.50692 0.0059 28	1.00000 0.0000 28	0.76463 0.0001 28	0.69824 0.0001 28	0.74394 0.0001 28	0.66249 0.0001 28	0.15168 0.4410 28	0.18926 0.3348 28	0.15353 0.4354 28	-0.01544 0.9379 28	-0.32877 0.0876 28
BO	0.64686 0.0002 28	0.59157 0.0009 28	0.76753 0.0001 28	0.76463 0.0001 28	1.00000 0.0000 28	0.45571 0.0148 28	0.71071 0.0001 28	0.48410 0.0090 28	-0.00991 0.9601 28	0.02665 0.8929 28	0.04835 0.8070 28	-0.05527 0.7800 28	-0.24607 0.2069 28
PBF	0.19471 0.3208 28	0.06759 0.7326 28	0.27670 0.1540 28	0.69824 0.0001 28	0.45571 0.0148 28	1.00000 0.0000 28	0.86841 0.0001 28	0.88081 0.0001 28	0.36839 0.0537 28	0.64672 0.0002 28	0.55523 0.0022 28	0.05644 0.7754 28	-0.05948 0.7637 28
BRF	0.37528 0.0491 28	0.29613 0.1260 28	0.58768 0.0010 28	0.74394 0.0001 28	0.71071 0.0001 28	0.86841 0.0001 28	1.00000 0.0000 28	0.86164 0.0001 28	0.18923 0.3348 28	0.48266 0.0093 28	0.42752 0.0232 28	0.08228 0.6772 28	-0.11217 0.5699 28
KF	0.24386 0.2111 28	0.09965 0.6139 28	0.39484 0.0376 28	0.66249 0.0001 28	0.48410 0.0090 28	0.88081 0.0001 28	0.86164 0.0001 28	1.00000 0.0000 28	0.21847 0.2640 28	0.66486 0.0001 28	0.57331 0.0014 28	0.04263 0.8295 28	0.04909 0.8041 28
ZNF	-0.14260 0.4691 28	-0.19311 0.3248 28	-0.18851 0.3367 28	0.15168 0.4410 28	-0.00991 0.9601 28	0.36839 0.0537 28	0.18923 0.3348 28	0.21847 0.2640 28	1.00000 0.0000 28	0.39592 0.0370 28	0.11300 0.5670 28	0.76033 0.0001 28	0.22685 0.2457 28
FEF	-0.11611 0.5563 28	-0.18015 0.3590 28	0.04107 0.8356 28	0.18926 0.3348 28	0.02665 0.8929 28	0.64672 0.0002 28	0.48266 0.0093 28	0.66486 0.0001 28	0.39592 0.0370 28	1.00000 0.0000 28	0.91240 0.0001 28	0.19875 0.3106 28	0.33503 0.0814 28
SIF	-0.11611 0.5563 28	-0.16029 0.4152 28	0.14089 0.4745 28	0.15353 0.4354 28	0.04835 0.8070 28	0.55523 0.0022 28	0.42752 0.0232 28	0.57331 0.0014 28	0.11300 0.5670 28	0.91240 0.0001 28	1.00000 0.0000 28	-0.10137 0.6078 28	0.24003 0.2186 28
CLF	-0.19705 0.3149 28	-0.12647 0.5213 28	-0.08588 0.6639 28	-0.01544 0.9379 28	-0.05527 0.7800 28	0.05644 0.7754 28	0.08228 0.6772 28	0.04263 0.8295 28	0.76033 0.0001 28	0.19875 0.3106 28	-0.10137 0.6078 28	1.00000 0.0000 28	0.15942 0.4178 28
NIF	-0.32613 0.0903 28	-0.38751 0.0416 28	-0.19215 0.3273 28	-0.32877 0.0876 28	-0.24607 0.2069 28	-0.05948 0.7637 28	-0.11217 0.5699 28	0.04909 0.8041 28	0.22685 0.2457 28	0.33503 0.0814 28	0.24003 0.2186 28	0.15942 0.4178 28	1.00000 0.0000 28

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
SF	0.22958	0.28128	0.53563	0.20299	0.44635	0.09855	0.32442	0.02425	0.00860	-0.14670	-0.04564	-0.03714	-0.02396
	0.2399	0.1471	0.0033	0.3002	0.0173	0.6178	0.0921	0.9025	0.9654	0.4563	0.8176	0.8512	0.9037
	28	28	28	28	28	28	28	28	28	28	28	28	28
NO3	0.55929	0.57045	0.65752	0.44922	0.64260	0 23458	0.49673	0.32987	0.22713	0.12396	0.06725	0.25440	-0.22887
	0.0024	0.0019	0.0002	0.0187	0.0003	0.2389	0.0084	0.0929	0.2546	0.5379	0.7389	0.2003	0.2508
	27	27	27	27	27	27	27	27	27	27	27	27	27
CO	0.52641	0.41998	0.31521	0.65324	0.54453	0.69311	0.71618	0.57225	0.26706	0.32003	0.21623	0.05800	0.47369
	0.0142	0.0580	0.1640	0.0013	0.0107	0.0005	0.0003	0.0067	0.2419	0.1573	0.3465	0.8028	0.0301
	21	21	21	21	21	21	21	21	21	21	21	21	21
NO	0.49046	0.49033	0.46283	0.11591	0.44772	0.20150	0.52331	0.24250	-0.07607	0.00739	0.03584	-0.09097	0.15638
	0.0150	0.0150	0.0228	0.5896	0.0282	0.3451	0.0087	0.2536	0.7239	0.9727	0.8679	0.6725	0.4656
	24	24	24	24	24	24	24	24	24	24	24	24	24
NO2	0.23577	0.12295	0.18784	0.51300	0.25803	0.81394	0.60462	0.63894	0.49678	0.66543	0.56268	0.14308	-0.00258
	0.2364	0.5412	0.3481	0.0062	0.1938	0.0001	0.0008	0.0003	0.0084	0.0001	0.0023	0.4765	0.9898
	27	27	27	27	27	27	27	27	27	27	27	27	27
O3	-0.29760	-0.35637	-0.63102	-0.03485	-0.47831	0.22480	-0.10942	0.10417	0.22249	0.34560	0.26416	-0.04431	0.02910
	0.1317	0.0681	0.0004	0.8630	0.0116	0.2596	0.5869	0.6050	0.2647	0.0774	0.1830	0.8263	0.8854
	27	27	27	27	27	27	27	27	27	27	27	27	27
SO2	-0.03006	0.00996	0.23266	-0.32781	0.00434	-0.10821	0.08671	-0.02683	-0.05085	0.02818	0.05407	0.01310	0.16543
	0.8817	0.9607	0.2429	0.0951	0.9829	0.5911	0.6671	0.8943	0.8011	0.8890	0.7888	0.9483	0.4096
	27	27	27	27	27	27	27	27	27	27	27	27	27

	SF	NO3	CO	NO	NO2	O3	SO2
TA98P	0.22958	0.55929	0.52641	0.49046	0.23577	-0.29760	-0.03006
	0.2399	0.0024	0.0142	0.0150	0.2364	0.1317	0.8817
	28	27	21	24	27	27	27
TA98M	0.28128	0.57045	0.41998	0.49033	0.12295	-0.35637	0.00996
	0.1471	0.0019	0.0580	0.0150	0.5412	0.0681	0.9607
	28	27	21	24	27	27	27
BAP	0.53563	0.65752	0.31521	0.46283	0.18784	-0.63102	0.23266
	0.0033	0.0002	0.1640	0.0228	0.3481	0.0004	0.2429
	28	27	21	24	27	27	27
COR	0.20299	0.44922	0.65324	0.11591	0.51300	-0.03485	-0.32781
	0.3002	0.0187	0.0013	0.5896	0.0062	0.8630	0.0951
	28	27	21	24	27	27	27
BO	0.44635	0.64260	0.54453	0.44772	0.25803	-0.47831	0.00434
	0.0173	0.0003	0.0107	0.0282	0.1938	0.0116	0.9829
	28	27	21	24	27	27	27

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	SF	NO3	CO	NO	NO2	O3	SO2
PBF	0.09855	0.23458	0.69311	0.20150	0.81394	0.22480	-0.10821
	0.6178	0.2389	0.0005	0.3451	0.0001	0.2596	0.5911
	28	27	21	24	27	27	27
BRF	0.32442	0.49673	0.71618	0.52331	0.60462	-0.10942	0.08671
	0.0921	0.0084	0.0003	0.0087	0.0008	0.5869	0.6671
	28	27	21	24	27	27	27
KF	0.02425	0.32987	0.57225	0.24250	0.63894	0.10419	-0.02683
	0.9025	0.0929	0.0067	0.2536	0.0003	0.6050	0.8943
	28	27	21	24	27	27	27
ZNF	0.00860	0.22713	0.26706	-0.07607	0.49678	0.22249	-0.05085
	0.9654	0.2546	0.2419	0.7239	0.0084	0.2647	0.8011
	28	27	21	24	27	27	27
FEF	-0.14670	0.12396	0.32003	0.00739	0.66543	0.34560	0.02818
	0.4563	0.5379	0.1573	0.9727	0.0001	0.0774	0.8890
	28	27	21	24	27	27	27
SIF	-0.04564	0.06725	0.21623	-0.03584	0.56268	0.26416	0.05407
	0.8176	0.7389	0.3465	0.8679	0.0023	0.1830	0.7888
	28	27	21	24	27	27	27
CLF	-0.03714	0.25440	0.05800	-0.09097	0.14308	-0.04431	0.01310
	0.8512	0.2003	0.8028	0.6725	0.4765	0.8263	0.9483
	28	27	21	24	27	27	27
NIF	-0.02396	-0.22887	0.47369	0.15638	-0.00258	0.02910	0.16543
	0.9037	0.2508	0.0301	0.4656	0.9898	0.8854	0.4096
	28	27	21	24	27	27	27
SF	1.00000	0.20932	0.24609	0.26478	-0.00797	-0.46722	0.09280
	0.0000	0.2947	0.2822	0.2112	0.9685	0.0140	0.6452
	28	27	21	24	27	27	27
NO3	0.20932	1.00000	0.45740	0.54089	0.25211	-0.41312	0.31640
	0.2947	0.0000	0.0426	0.0077	0.2141	0.0359	0.1153
	27	27	20	23	26	26	26
CO	0.24609	0.45740	1.00000	0.82841	0.54243	0.01563	0.31080
	0.2822	0.0426	0.0000	0.0001	0.0111	0.9464	0.1703
	21	20	21	21	21	21	21
NO	0.26478	0.54089	0.82841	1.00000	0.13972	-0.33512	0.67986
	0.2112	0.0077	0.0001	0.0000	0.5149	0.1094	0.0003
	24	23	21	24	24	24	24
NO2	-0.00797	0.25211	0.54243	0.13972	1.00000	0.23446	-0.01640
	0.9685	0.2141	0.0111	0.5149	0.0000	0.2391	0.9353
	27	26	21	24	27	27	27

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	SF	NO3	CO	NO	NO2	O3	SO2
O3	-0.46722	-0.41312	0.01563	-0.33512	0.23446	1.00000	-0.41739
	0.0140	0.0359	0.9464	0.1094	0.2391	0.0000	0.0303
	27	26	21	24	27	27	27
SO2	0.09280	0.31640	0.31080	0.67986	-0.01640	-0.41739	1.00000
	0.6452	0.1153	0.1703	0.0003	0.9353	0.0303	0.0000
	27	26	21	24	27	27	27

VARIABLE	N	MEAN	STD DEV	SUM	MINIMUM	MAXIMUM
TA98P	44	18.45227271	10.76969236	811.89999920	0.60000000	43.19999993
TA98M	44	9.05227272	5.38060636	398.29999963	0.40000000	23.29999995
BAP	44	0.22045455	0.30160774	9.69999999	0.10000000	1.60000000
COR	44	0.50227273	0.37384777	22.09999999	0.10000000	1.60000000
BO	44	0.75454545	1.00381934	33.19999998	0.10000000	5.10000000
PBF	44	229.16954545	194.74778745	10083.46000000	17.00000000	950.00000000
BRF	44	43.51818182	37.41857065	1914.80000000	6.00000000	184.00000000
KF	44	131.34545455	93.36273211	5779.20000000	31.00000000	493.00000000
ZNF	44	20.22954545	15.20854380	890.10000000	3.00000000	74.00000000
FEF	44	118.57045455	105.55881548	5217.10000000	12.00000000	450.00000000
SIF	44	268.81818182	226.67652526	11828.00000000	64.00000000	880.00000000
CLF	44	360.98863636	634.90915161	15883.50000000	12.00000000	3050.00000000
NIF	44	7.61136364	8.33619778	334.89999998	2.00000000	38.50000000
SF	44	1796.75681818	1354.97035141	79057.30000000	442.00000000	6780.00000000
NO3	44	7.35909091	5.28205906	323.79999985	3.00000000	24.50000000
CO	33	1.02727254	0.50017025	33.89999372	0.19999999	2.79999924
NO	39	1.63589716	1.47067583	63.79998940	0	6.29999924
NO2	42	2.60238061	1.41309002	109.29998565	0.09999996	6.29999924
O3	42	1.42142833	1.19399613	59.69998980	0	4.39999962
SO2	42	0.33571422	0.67093710	14.09999716	0	3.79999924

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	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
TA98P	1.00000 0.0000 44	0.95237 0.0001 44	0.56104 0.0001 44	0.55072 0.0001 44	0.65764 0.0001 44	0.33045 0.0285 44	0.43091 0.0035 44	0.27040 0.0759 44	0.34223 0.0230 44	0.21529 0.1605 44	0.05318 0.7317 44	-0.30865 0.0415 44	0.10093 0.5145 44
TA98M	0.95237 0.0001 44	1.00000 0.0000 44	0.55764 0.0001 44	0.50459 0.0005 44	0.63421 0.0001 44	0.35047 0.0197 44	0.43332 0.0033 44	0.33195 0.0277 44	0.46691 0.0014 44	0.32433 0.0317 44	0.16138 0.2953 44	-0.27883 0.0668 44	0.18899 0.2192 44
BAP	0.56104 0.0001 44	0.55764 0.0001 44	1.00000 0.0000 44	0.46158 0.0016 44	0.96100 0.0001 44	0.06291 0.6850 44	0.28304 0.0626 44	0.13823 0.3709 44	0.09523 0.5386 44	-0.08884 0.5663 44	-0.12002 0.4377 44	0.00207 0.9894 44	-0.11692 0.4498 44
COR	0.55072 0.0001 44	0.50459 0.0005 44	0.46158 0.0016 44	1.00000 0.0000 44	0.53818 0.0002 44	0.73445 0.0001 44	0.76993 0.0001 44	0.52936 0.0002 44	0.27870 0.0670 44	0.32813 0.0297 44	0.09155 0.5545 44	-0.14512 0.3473 44	-0.12627 0.4141 44
BO	0.65764 0.0001 44	0.63421 0.0001 44	0.96100 0.0001 44	0.53818 0.0002 44	1.00000 0.0000 44	0.12564 0.4164 44	0.36631 0.0145 44	0.17730 0.2496 44	0.15212 0.3243 44	-0.07702 0.6192 44	-0.15497 0.3152 44	-0.05387 0.7284 44	-0.07080 0.6479 44
PBF	0.33045 0.0285 44	0.35047 0.0197 44	0.06291 0.6850 44	0.73445 0.0001 44	0.12564 0.4164 44	1.00000 0.0000 44	0.91786 0.0001 44	0.84828 0.0001 44	0.63036 0.0001 44	0.77583 0.0001 44	0.51462 0.0004 44	-0.18020 0.2418 44	0.32380 0.0320 44
BRF	0.43091 0.0035 44	0.43332 0.0033 44	0.28304 0.0626 44	0.76993 0.0001 44	0.36631 0.0145 44	0.91786 0.0001 44	1.00000 0.0000 44	0.84340 0.0001 44	0.65557 0.0001 44	0.65958 0.0001 44	0.40304 0.0067 44	-0.03362 0.8285 44	0.29442 0.0524 44
KF	0.27040 0.0759 44	0.33195 0.0277 44	0.13823 0.3709 44	0.52936 0.0002 44	0.17730 0.2496 44	0.84828 0.0001 44	0.84340 0.0001 44	1.00000 0.0000 44	0.70326 0.0001 44	0.73538 0.0001 44	0.55996 0.0001 44	0.01384 0.9289 44	0.35888 0.0167 44
ZNF	0.34223 0.0230 44	0.46691 0.0014 44	0.09523 0.5386 44	0.27870 0.0670 44	0.15212 0.3243 44	0.63036 0.0001 44	0.65557 0.0001 44	0.70326 0.0001 44	1.00000 0.0000 44	0.65550 0.0001 44	0.50242 0.0005 44	-0.08205 0.5965 44	0.39785 0.0075 44
FEF	0.21529 0.1605 44	0.32433 0.0317 44	-0.08884 0.5663 44	0.32813 0.0297 44	-0.07702 0.6192 44	0.77583 0.0001 44	0.65958 0.0001 44	0.73538 0.0001 44	0.65550 0.0001 44	1.00000 0.0000 44	0.86213 0.0001 44	-0.20812 0.1752 44	0.52012 0.0003 44
SIF	0.05318 0.7317 44	0.16138 0.2953 44	-0.12002 0.4377 44	0.09155 0.5545 44	-0.15497 0.3152 44	0.51462 0.0004 44	0.40304 0.0067 44	0.55996 0.0001 44	0.50242 0.0005 44	0.86213 0.0001 44	1.00000 0.0000 44	-0.10747 0.4874 44	0.48434 0.0009 44
CLF	-0.30865 0.0415 44	-0.27883 0.0668 44	0.00207 0.9894 44	-0.14512 0.3473 44	-0.05387 0.7284 44	-0.18020 0.2418 44	-0.03362 0.8285 44	0.01384 0.9289 44	-0.08205 0.5965 44	-0.20812 0.1752 44	-0.10747 0.4874 44	1.00000 0.0000 44	-0.16495 0.2846 44
NIF	0.10093 0.5145 44	0.18899 0.2192 44	-0.11692 0.4498 44	-0.12627 0.4141 44	-0.07080 0.6479 44	0.32380 0.0320 44	0.29442 0.0524 44	0.35888 0.0167 44	0.39785 0.0075 44	0.52012 0.0003 44	0.48434 0.0009 44	-0.16495 0.2846 44	1.00000 0.0000 44

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	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
SF	0.21660	0.20210	0.25827	-0.08092	0.34611	0.00377	0.20229	0.25261	0.29022	-0.03143	-0.02922	-0.03505	0.37826
	0.1579	0.1883	0.0905	0.6016	0.0214	0.9806	0.1879	0.0981	0.0560	0.8395	0.8507	0.8213	0.0114
	44	44	44	44	44	44	44	44	44	44	44	44	44
NO3	0.49232	0.50623	0.29892	0.00287	0.44009	-0.12038	0.05446	0.07791	0.23518	-0.17030	-0.15959	-0.04835	0.17795
	0.0007	0.0005	0.0487	0.9852	0.0028	0.4363	0.7255	0.6152	0.1244	0.2691	0.3008	0.7553	0.2478
	44	44	44	44	44	44	44	44	44	44	44	44	44
CO	0.54987	0.57589	0.27606	0.72226	0.33797	0.60931	0.65025	0.47550	0.36785	0.43089	0.17300	-0.04732	0.15085
	0.0009	0.0005	0.1199	0.0001	0.0544	0.0002	0.0001	0.0052	0.0352	0.0123	0.3357	0.7937	0.4021
	33	33	33	33	33	33	33	33	33	33	33	33	33
NO	0.50903	0.41587	0.38206	0.70751	0.46865	0.61193	0.71055	0.49081	0.28361	0.39356	0.27777	-0.15733	0.15573
	0.0009	0.0085	0.0164	0.0001	0.0026	0.0001	0.0001	0.0015	0.0802	0.0132	0.0869	0.3388	0.3438
	39	39	39	39	39	39	39	39	39	39	39	39	39
NO2	0.39623	0.42635	0.09859	0.70120	0.15866	0.83130	0.73349	0.62731	0.46326	0.63064	0.43895	-0.18107	0.39652
	0.0094	0.0049	0.5345	0.0001	0.3156	0.0001	0.0001	0.0001	0.0020	0.0001	0.0036	0.2512	0.0093
	42	42	42	42	42	42	42	42	42	42	42	42	42
O3	-0.50293	-0.47109	-0.41254	-0.50418	-0.52620	-0.19516	-0.35514	-0.04447	-0.30810	0.08825	0.14842	0.04660	0.11746
	0.0007	0.0016	0.0066	0.0007	0.0003	0.2155	0.0210	0.7797	0.0471	0.5784	0.3649	0.7695	0.4588
	42	42	42	42	42	42	42	42	42	42	42	42	42
SO2	0.09780	0.09423	-0.08735	-0.27067	-0.03854	-0.07616	0.05639	0.05851	0.14078	0.09035	0.17755	-0.04330	0.49577
	0.5378	0.5528	0.5823	0.0830	0.8086	0.6317	0.7228	0.7128	0.3739	0.5693	0.2606	0.7854	0.0008
	42	42	42	42	42	42	42	42	42	42	42	42	42

	SF	NO3	CO	NO	NO2	O3	SO2
TA98P	0.21660	0.49232	0.54987	0.50903	0.39623	-0.50293	0.09780
	0.1579	0.0007	0.0009	0.0009	0.0094	0.0007	0.5378
	44	44	33	39	42	42	42
TA98M	0.20210	0.50623	0.57589	0.41587	0.42635	-0.47109	0.09423
	0.1883	0.0005	0.0005	0.0085	0.0049	0.0016	0.5528
	44	44	33	39	42	42	42
BAP	0.25827	0.29892	0.27606	0.38206	0.09859	-0.41254	-0.08735
	0.0905	0.0487	0.1199	0.0164	0.5345	0.0066	0.5823
	44	44	33	39	42	42	42
COR	-0.08092	0.00287	0.72226	0.70751	0.70120	-0.50418	-0.27067
	0.6016	0.9852	0.0001	0.0001	0.0001	0.0007	0.0830
	44	44	33	39	42	42	42
BO	0.34611	0.44009	0.33797	0.46865	0.15866	-0.52620	-0.03854
	0.0214	0.0028	0.0544	0.0026	0.3156	0.0003	0.8086
	44	44	33	39	42	42	42

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	SF	NO3	CO	NO	NO2	O3	SO2
PBF	0.00377	-0.12038	0.60931	0.61193	0.83130	-0.19516	-0.07616
	0.9806	0.4363	0.0002	0.0001	0.0001	0.2155	0.6317
	44	44	33	39	42	42	42
BRF	0.20229	0.05446	0.65025	0.71055	0.73349	-0.35514	0.05639
	0.1879	0.7255	0.0001	0.0001	0.0001	0.0210	0.7228
	44	44	33	39	42	42	42
KF	0.25261	0.07791	0.47550	0.49081	0.62731	-0.04447	0.05851
	0.0981	0.6152	0.0052	0.0015	0.0001	0.7797	0.7128
	44	44	33	39	42	42	42
ZNF	0.29022	0.23518	0.36785	0.28361	0.46326	-0.30810	0.14078
	0.0560	0.1244	0.0352	0.0802	0.0020	0.0471	0.3739
	44	44	33	39	42	42	42
FEF	-0.03143	-0.17030	0.43089	0.39356	0.63064	0.08825	0.09035
	0.8395	0.2691	0.0123	0.0132	0.0001	0.5784	0.5693
	44	44	33	39	42	42	42
SIF	-0.02922	-0.15959	0.17300	-0.27777	0.43895	0.14342	0.17755
	0.8507	0.3008	0.3357	0.0869	0.0036	0.3649	0.2606
	44	44	33	39	42	42	42
CLF	-0.03505	-0.04835	-0.04732	-0.15733	-0.18107	0.04660	-0.04330
	0.8213	0.7553	0.7937	0.3388	0.2512	0.7695	0.7854
	44	44	33	39	42	42	42
NIF	0.37826	0.17795	0.15085	0.15573	0.39652	0.11746	0.49577
	0.0114	0.2478	0.4021	0.3438	0.0093	0.4588	0.0008
	44	44	33	39	42	42	42
SF	1.00000	0.57074	-0.00124	0.03909	-0.02841	-0.17580	0.47123
	0.0000	0.0001	0.9945	0.8132	0.8583	0.2654	0.0016
	44	44	33	39	42	42	42
NO3	0.57074	1.00000	0.12482	0.01750	-0.11774	-0.36096	0.45251
	0.0001	0.0000	0.4889	0.9158	0.4577	0.0189	0.0026
	44	44	33	39	42	42	42
CO	-0.00124	0.12482	1.00000	0.62874	0.57099	-0.36728	0.02018
	0.9945	0.4889	0.0000	0.0001	0.0005	0.0355	0.9113
	33	33	33	33	33	33	33
NO	0.03909	0.01750	0.62874	1.00000	0.54775	-0.44879	0.13543
	0.8132	0.9158	0.0001	0.0000	0.0003	0.0042	0.4110
	39	39	33	39	39	39	39
NO2	-0.02841	-0.11774	0.57099	0.54775	1.00000	-0.28539	-0.02710
	0.8583	0.4577	0.0005	0.0003	0.0000	0.0669	0.8647
	42	42	33	39	42	42	42

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	SF	NO3	CO	NO	NO2	O3	SO2
O3	-0.17580	-0.36096	-0.36728	-0.44879	-0.28539	1.00000	-0.07679
	0.2654	0.0189	0.0355	0.0042	0.0669	0.0000	0.6288
	42	42	33	39	42	42	42
SO2	0.47123	0.45251	0.02018	0.13543	-0.02710	-0.07679	1.00000
	0.0016	0.0026	0.9113	0.4110	0.8647	0.6288	0.0000
	42	42	33	39	42	42	42

VARIABLE	N	MEAN	STD DEV	SUM	MINIMUM	MAXIMUM
TA98P	18	22.41111109	9.49122745	403.39999966	7.80000000	41.00000000
TA98M	18	10.34999999	5.24182836	186.29999981	3.40000000	23.29999995
BAP	18	0.15000000	0.11504475	2.70000000	0.10000000	0.50000000
COR	18	0.52222222	0.39490215	9.39999999	0.10000000	1.60000000
BO	18	0.60000000	0.59606553	10.80000000	0.10000000	2.00000000
PBF	18	251.66666667	208.44607146	4530.00000000	78.00000000	950.00000000
BRF	18	42.83333333	41.58089210	771.00000000	12.00000000	184.00000000
KF	18	157.94444444	126.92029561	2843.00000000	48.00000000	491.00000000
ZNF	18	30.72222222	14.74012354	553.00000000	17.00000000	67.00000000
FEF	18	156.55555556	110.37381521	2818.00000000	23.00000000	450.00000000
SIF	18	338.33333333	211.55502691	6090.00000000	111.00000000	770.00000000
CLF	18	207.11111111	327.61919948	3728.00000000	12.00000000	1450.00000000
NIF	18	5.33333333	3.30774492	96.00000000	2.00000000	14.00000000
SF	18	1450.94444444	638.56488658	26117.00000000	442.00000000	2680.00000000
NO3	18	8.30000000	5.45581179	149.39999992	3.50000000	24.09999996
CO	18	0.94444434	0.58933865	16.99999809	0.29999995	2.79999924
NO	18	1.28888869	1.13131291	23.19999641	0	4.00000000
NO2	18	2.30555516	1.31930107	41.49999291	0.09999996	6.29999924
O3	18	2.32777747	1.67746482	41.89999443	0	5.89999962
SO2	18	0.12222221	0.17339364	2.19999981	0	0.59999996

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CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / N = 18

	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
TA98P	1.00000 0.0000	0.91598 0.0001	0.68417 0.0017	0.63507 0.0046	0.70777 0.0010	0.45839 0.0557	0.56313 0.0150	0.48713 0.0403	0.19524 0.4375	0.35988 0.1424	0.17180 0.4955	-0.31142 0.2084	0.72874 0.0006
TA98M	0.91598 0.0001	1.00000 0.0000	0.74963 0.0003	0.56550 0.0145	0.71316 0.0009	0.41446 0.0873	0.54966 0.0181	0.40132 0.0988	0.30860 0.2128	0.40986 0.0912	0.18496 0.4625	-0.15585 0.5369	0.64087 0.0042
BAP	0.68417 0.0017	0.74963 0.0003	1.00000 0.0000	0.82866 0.0001	0.84923 0.0001	0.63630 0.0045	0.72735 0.0006	0.49209 0.0380	0.41453 0.0872	0.40720 0.0935	0.01378 0.9567	-0.07288 0.7738	0.49465 0.0369
COR	0.63507 0.0046	0.56550 0.0145	0.82866 0.0001	1.00000 0.0000	0.84966 0.0001	0.82039 0.0001	0.87254 0.0001	0.75878 0.0003	0.37503 0.1252	0.49121 0.0384	0.12672 0.6163	-0.21349 0.3950	0.57042 0.0134
BO	0.70777 0.0010	0.71316 0.0009	0.84923 0.0001	0.84966 0.0001	1.00000 0.0000	0.54649 0.0189	0.68519 0.0017	0.55828 0.0160	0.34814 0.1568	0.17373 0.4906	-0.21593 0.3895	-0.05853 0.8176	0.55791 0.0161
PBF	0.45839 0.0557	0.41446 0.0873	0.63630 0.0045	0.82039 0.0001	0.54649 0.0189	1.00000 0.0000	0.97218 0.0001	0.82598 0.0001	0.47157 0.0482	0.74449 0.0004	0.33422 0.1752	-0.22037 0.3796	0.66759 0.0025
BRF	0.56313 0.0150	0.54966 0.0181	0.72735 0.0006	0.87254 0.0001	0.68519 0.0017	0.97218 0.0001	1.00000 0.0000	0.87204 0.0001	0.46741 0.0505	0.69153 0.0015	0.27482 0.2697	-0.14037 0.5785	0.68301 0.0018
KF	0.48713 0.0403	0.40132 0.0988	0.49209 0.0380	0.75878 0.0003	0.55828 0.0160	0.82598 0.0001	0.87204 0.0001	1.00000 0.0000	0.28700 0.2482	0.63164 0.0049	0.41478 0.0870	-0.13133 0.6035	0.59330 0.0094
ZNF	0.19524 0.4375	0.30860 0.2128	0.41453 0.0872	0.37503 0.1252	0.34814 0.1568	0.47157 0.0482	0.46741 0.0505	0.28700 0.2482	1.00000 0.0000	0.26191 0.2938	-0.04128 0.8708	-0.12697 0.6156	0.33862 0.1693
FEF	0.35988 0.1424	0.40986 0.0912	0.40720 0.0935	0.49121 0.0384	0.17373 0.4906	0.74449 0.0004	0.69153 0.0015	0.63164 0.0049	0.26191 0.2938	1.00000 0.0000	0.82141 0.0001	-0.35441 0.1490	0.51473 0.0288
SIF	0.17180 0.4955	0.18496 0.4625	0.01378 0.9567	0.12672 0.6163	-0.21593 0.3895	0.33422 0.1752	0.27482 0.2697	0.41478 0.0870	-0.04128 0.8708	0.82141 0.0001	1.00000 0.0000	-0.33167 0.1788	0.19805 0.4308
CLF	-0.31142 0.2084	-0.15585 0.5369	-0.07288 0.7738	-0.21349 0.3950	-0.05853 0.8176	-0.22037 0.3796	-0.14037 0.5785	-0.13133 0.6035	-0.12697 0.6156	-0.35441 0.1490	-0.33167 0.1788	1.00000 0.0000	-0.25158 0.3139
NIF	0.72874 0.0006	0.64087 0.0042	0.49465 0.0369	0.57042 0.0134	0.55791 0.0161	0.66759 0.0025	0.68301 0.0018	0.59330 0.0094	0.33862 0.1693	0.51473 0.0288	0.19805 0.4308	-0.25158 0.3139	1.00000 0.0000
SF	0.28894 0.2449	0.31765 0.1990	0.24930 0.3185	0.18004 0.4747	0.30830 0.2132	0.16776 0.5058	0.21522 0.3911	0.16711 0.5075	0.63384 0.0047	-0.12749 0.6142	-0.23080 0.3568	-0.17360 0.4909	0.21072 0.4013
NO3	0.55567 0.0167	0.61048 0.0071	0.27834 0.2634	0.20313 0.4188	0.52764 0.0244	-0.12527 0.6204	0.03163 0.9008	0.03703 0.8840	0.10979 0.6645	-0.26820 0.2819	-0.26761 0.2830	0.09971 0.6938	0.33508 0.1741
CO	0.70534 0.0011	0.70511 0.0011	0.72011 0.0008	0.77904 0.0001	0.65976 0.0029	0.77245 0.0002	0.81431 0.0001	0.68611 0.0017	0.69558 0.0013	0.59428 0.0093	0.27833 0.2634	-0.21896 0.3827	0.69202 0.0015
NO	0.58992 0.0100	0.50579 0.0322	0.55139 0.0177	0.56017 0.0156	0.42220 0.0809	0.68843 0.0016	0.70372 0.0011	0.67002 0.0023	0.37301 0.1274	0.62731 0.0053	0.34285 0.1637	-0.40289 0.0974	0.58109 0.0114

STATION=7430

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / N = 18

	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
NO2	0.25001	0.22673	0.58716	0.74041	0.49444	0.83711	0.76285	0.45880	0.42236	0.53119	0.06999	-0.20935	0.50099
	0.3170	0.3656	0.0104	0.0004	0.0370	0.0001	0.0002	0.0555	0.0808	0.0233	0.7826	0.4044	0.0342
O3	-0.33221	-0.49548	-0.38254	-0.12175	-0.45888	-0.00093	-0.13765	0.12685	-0.18999	0.23454	0.45794	-0.20550	-0.06538
	0.1780	0.0365	0.1172	0.6303	0.0554	0.9971	0.5860	0.6160	0.4502	0.3489	0.0560	0.4133	0.7966
SO2	-0.25143	-0.25953	-0.32437	-0.26536	-0.50654	-0.01752	-0.13897	-0.03603	0.10613	0.28547	0.48439	-0.21512	-0.20854
	0.3142	0.2983	0.1891	0.2872	0.0319	0.9450	0.5823	0.8871	0.6751	0.2508	0.0416	0.3913	0.4063

	SF	NO3	CO	NO	NO2	O3	SO2
TA98P	0.28894	0.55567	0.70534	0.58992	0.25001	-0.33221	-0.25143
	0.2449	0.0167	0.0011	0.0100	0.3170	0.1780	0.3142
TA98M	0.31765	0.61048	0.70511	0.50579	0.22673	-0.49548	-0.25953
	0.1990	0.0071	0.0011	0.0322	0.3656	0.0365	0.2983
BAP	0.24930	0.27834	0.72011	0.55139	0.58716	-0.38254	-0.32437
	0.3185	0.2634	0.0008	0.0177	0.0104	0.1172	0.1891
COR	0.18004	0.20313	0.77904	0.56017	0.74041	-0.12175	-0.26536
	0.4747	0.4188	0.0001	0.0156	0.0004	0.6303	0.2872
BO	0.30830	0.52764	0.65976	0.42220	0.49444	-0.45888	-0.50654
	0.2132	0.0244	0.0029	0.0809	0.0370	0.0554	0.0319
PBF	0.16776	-0.12527	0.77245	0.68843	0.83711	-0.00093	-0.01752
	0.5058	0.6204	0.0002	0.0016	0.0001	0.9971	0.9450
BRF	0.21522	0.03163	0.81431	0.70372	0.76285	-0.13765	-0.13897
	0.3911	0.9008	0.0001	0.0011	0.0002	0.5860	0.5823
KF	0.16711	0.03703	0.68611	0.67002	0.45880	0.12685	-0.03603
	0.5075	0.8840	0.0017	0.0023	0.0555	0.6160	0.8871
ZNF	0.63384	0.10979	0.69558	0.37301	0.42236	-0.18999	0.10613
	0.0047	0.6645	0.0013	0.1274	0.0808	0.4502	0.6751
FEF	-0.12749	-0.26820	0.59428	0.62731	0.53119	0.23454	0.28547
	0.6142	0.2819	0.0093	0.0053	0.0233	0.3489	0.2508
SIF	-0.23080	-0.26761	0.27833	0.34285	0.06999	0.45794	0.48439
	0.3568	0.2830	0.2634	0.1637	0.7826	0.0560	0.0416
CLF	-0.17360	0.09971	-0.21896	-0.40289	-0.20935	-0.20550	-0.21512
	0.4909	0.6938	0.3827	0.0974	0.4044	0.4133	0.3913
NIF	0.21072	0.33508	0.69202	0.58109	0.50099	-0.06538	-0.20854
	0.4013	0.1741	0.0015	0.0114	0.0342	0.7966	0.4063

CORRELATION COEFFICIENTS / PROB >|R| UNDER H0:RHO=0 / N = 18

	SF	NO3	CO	NO	NO2	O3	SO2
SF	1.00000	0.46453	0.37823	0.07208	0.06238	-0.31989	0.01000
	0.0000	0.0521	0.1217	0.7762	0.8058	0.1956	0.9686
NO3	0.46453	1.00000	0.24259	-0.18670	-0.14874	-0.45076	-0.36811
	0.0521	0.0000	0.3321	0.4582	0.5558	0.0605	0.1328
CO	0.37823	0.24259	1.00000	0.71631	0.54136	-0.14710	0.01855
	0.1217	0.3321	0.0000	0.0008	0.0203	0.5602	0.9418
NO	0.07208	-0.18670	0.71631	1.00000	0.34371	0.02900	0.04331
	0.7762	0.4582	0.0008	0.0000	0.1626	0.9091	0.8645
NO2	0.06238	-0.14874	0.54136	0.34371	1.00000	-0.07769	-0.12657
	0.8058	0.5558	0.0203	0.1626	0.0000	0.7593	0.6167
O3	-0.31989	-0.45076	-0.14710	0.02900	-0.07769	1.00000	0.50537
	0.1956	0.0605	0.5602	0.9091	0.7593	0.0000	0.0324
SO2	0.01000	-0.36811	0.01855	0.04331	-0.12657	0.50537	1.00000
	0.9686	0.1328	0.9418	0.8645	0.6167	0.0324	0.0000

STATION=7433

VARIABLE	N	MEAN	STD DEV	SUM	MINIMUM	MAXIMUM
TA98P	18	15.53333332	11.60724924	279.59999980	1.70000000	38.00000000
TA98M	18	6.72222222	4.76526100	124.59999995	1.60000000	20.00000000
BAP	18	0.18888889	0.17111705	3.40000000	0.10000000	0.70000000
COR	18	0.60555556	0.38267206	10.89999999	0.10000000	1.60000000
BO	18	0.75555556	0.80163232	13.59999999	0.10000000	2.50000000
PBF	18	191.83333333	142.44844269	3453.00000000	49.00000000	576.00000000
BRF	18	38.72222222	26.73135536	697.00000000	7.00000000	84.00000000
KF	18	116.11111111	77.57947643	2090.00000000	31.00000000	349.00000000
ZNF	18	23.16666667	29.35432627	417.00000000	3.00000000	123.00000000
FEF	18	77.33333333	76.15309657	1392.00000000	12.00000000	290.00000000
SIF	18	138.61111111	133.89334601	2495.00000000	24.00000000	654.00000000
CLF	18	556.33333333	921.65918162	10014.00000000	12.00000000	3050.00000000
NIF	18	6.33333333	5.58358940	114.00000000	2.00000000	20.00000000
SF	18	2409.27777778	1960.08414602	43367.00000000	781.00000000	7640.00000000
NO3	17	7.20588235	4.09946194	122.49999993	3.50000000	16.29999995
CO	18	1.15555547	0.36817857	20.79999846	0.50000000	2.09999943
NO	18	1.34999975	1.20257059	24.29999548	0	3.50000000
NO2	18	2.21111060	1.12453320	39.79999083	0.69999999	5.29999924
O3	18	2.22222193	1.29685385	39.99999470	0.09999996	4.39999962
SO2	18	0.12222221	0.18959881	2.19999987	0	0.50000000

CORRELATIONS OF RICHMOND EPISODE DATA, pp. 20-24

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
TA98P	1.00000 0.0000 18	0.92786 0.0001 18	0.69499 0.0014 18	0.74542 0.0004 18	0.87202 0.0001 18	0.57443 0.0127 18	0.80267 0.0001 18	0.51850 0.0275 18	0.30529 0.2180 18	0.38214 0.1176 18	0.52537 0.0252 18	-0.28647 0.2491 18	0.29335 0.2374 18
TA98M	0.92786 0.0001 18	1.00000 0.0000 18	0.72027 0.0007 18	0.55831 0.0160 18	0.82658 0.0001 18	0.36347 0.1382 18	0.72067 0.0007 18	0.30814 0.2135 18	0.19909 0.4284 18	0.18381 0.4653 18	0.27021 0.2782 18	-0.24485 0.3275 18	0.16441 0.5145 18
BAP	0.69499 0.0014 18	0.72027 0.0007 18	1.00000 0.0000 18	0.26151 0.2945 18	0.90959 0.0001 18	0.02019 0.9366 18	0.41595 0.0860 18	0.08606 0.7342 18	-0.03357 0.8948 18	-0.09269 0.7145 18	0.06630 0.7938 18	-0.11526 0.6488 18	-0.03899 0.8779 18
COR	0.74542 0.0004 18	0.55831 0.0160 18	0.26151 0.2945 18	1.00000 0.0000 18	0.55119 0.0177 18	0.80568 0.0001 18	0.79948 0.0001 18	0.64018 0.0042 18	0.55552 0.0167 18	0.63819 0.0044 18	0.71161 0.0009 18	-0.17908 0.4771 18	0.20556 0.4132 18
BO	0.87202 0.0001 18	0.82658 0.0001 18	0.90959 0.0001 18	0.55119 0.0177 18	1.00000 0.0000 18	0.30154 0.2240 18	0.63241 0.0049 18	0.29046 0.2423 18	0.14882 0.5556 18	0.16657 0.5089 18	0.29835 0.2292 18	-0.22686 0.3653 18	0.12310 0.6265 18
PBF	0.57443 0.0127 18	0.36347 0.1382 18	0.02019 0.9366 18	0.80568 0.0001 18	0.30154 0.2240 18	1.00000 0.0000 18	0.81421 0.0001 18	0.87240 0.0001 18	0.70828 0.0010 18	0.83543 0.0001 18	0.79259 0.0001 18	-0.10927 0.6660 18	0.58337 0.0110 18
BRF	0.80267 0.0001 18	0.72067 0.0007 18	0.41595 0.0860 18	0.79948 0.0001 18	0.63241 0.0049 18	0.81421 0.0001 18	1.00000 0.0000 18	0.72937 0.0006 18	0.59161 0.0097 18	0.60433 0.0079 18	0.58652 0.0105 18	0.07669 0.7623 18	0.34274 0.1638 18
KF	0.51850 0.0275 18	0.30814 0.2135 18	0.08606 0.7342 18	0.64018 0.0042 18	0.29046 0.2423 18	0.87240 0.0001 18	0.72937 0.0006 18	1.00000 0.0000 18	0.70005 0.0012 18	0.87520 0.0001 18	0.85297 0.0001 18	0.12863 0.6110 18	0.58343 0.0110 18
ZNF	0.30529 0.2180 18	0.19909 0.4284 18	-0.03357 0.8948 18	0.55552 0.0167 18	0.14882 0.5556 18	0.70828 0.0010 18	0.59161 0.0097 18	0.70005 0.0012 18	1.00000 0.0000 18	0.87145 0.0001 18	0.54483 0.0194 18	0.11953 0.6366 18	0.54444 0.0195 18
FEF	0.38214 0.1176 18	0.18381 0.4653 18	-0.09269 0.7145 18	0.63819 0.0044 18	0.16657 0.5089 18	0.83543 0.0001 18	0.60433 0.0079 18	0.87520 0.0001 18	0.87145 0.0001 18	1.00000 0.0000 18	0.78481 0.0001 18	-0.00453 0.9858 18	0.61174 0.0070 18
SIF	0.52537 0.0252 18	0.27021 0.2782 18	0.06630 0.7938 18	0.71161 0.0009 18	0.29835 0.2292 18	0.79259 0.0001 18	0.58652 0.0105 18	0.85297 0.0001 18	0.54483 0.0194 18	0.78481 0.0001 18	1.00000 0.0000 18	-0.07886 0.7558 18	0.51571 0.0285 18
CLF	-0.28647 0.2491 18	-0.24485 0.3275 18	-0.11526 0.6488 18	-0.17908 0.4771 18	-0.22686 0.3653 18	-0.10927 0.6660 18	0.07669 0.7623 18	0.12863 0.6110 18	0.11953 0.6366 18	-0.00453 0.9858 18	-0.07886 0.7558 18	1.00000 0.0000 18	-0.20751 0.4087 18
NIF	0.29335 0.2374 18	0.16441 0.5145 18	-0.03899 0.8779 18	0.20556 0.4132 18	0.12310 0.6265 18	0.58337 0.0110 18	0.34274 0.1638 18	0.58343 0.0110 18	0.54444 0.0195 18	0.61174 0.0070 18	0.51571 0.0285 18	-0.20751 0.4087 18	1.00000 0.0000 18

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	BIF	CLF	NIF
SF	0.71313	0.83969	0.71558	0.22603	0.72637	0.09862	0.44639	0.10128	-0.05963	-0.05255	0.12180	-0.30267	0.22105
	0.0009	0.0001	0.0008	0.3671	0.0006	0.6970	0.0633	0.6892	0.8142	0.8359	0.6302	0.2222	0.3781
	18	18	18	18	18	18	18	18	18	18	18	18	18
NO3	0.70324	0.68520	0.57713	0.37124	0.69133	0.40748	0.55993	0.54401	0.50399	0.50803	0.47233	-0.14438	0.61873
	0.0016	0.0024	0.0153	0.1423	0.0021	0.1045	0.0194	0.0240	0.0391	0.0373	0.0556	0.5804	0.0081
	17	17	17	17	17	17	17	17	17	17	17	17	17
CO	0.66396	0.50083	0.22512	0.86192	0.43936	0.76253	0.78881	0.72386	0.60760	0.67066	0.69959	0.00752	0.11923
	0.0027	0.0343	0.3691	0.0001	0.0681	0.0002	0.0001	0.0007	0.0075	0.0023	0.0012	0.9764	0.6375
	18	18	18	18	18	18	18	18	18	18	18	18	18
NO	0.83251	0.77972	0.67176	0.75225	0.77128	0.57718	0.87660	0.45750	0.37468	0.31146	0.37828	-0.03706	0.02979
	0.0001	0.0001	0.0023	0.0003	0.0002	0.0121	0.0001	0.0563	0.1255	0.2083	0.1217	0.8839	0.9066
	18	18	18	18	18	18	18	18	18	18	18	18	18
NO2	0.59259	0.39963	0.16270	0.67239	0.35817	0.89205	0.78246	0.92474	0.74285	0.87231	0.80553	0.05950	0.64767
	0.0096	0.1004	0.5189	0.0022	0.1444	0.0001	0.0001	0.0001	0.0004	0.0001	0.0001	0.8146	0.0037
	18	18	18	18	18	18	18	18	18	18	18	18	18
O3	-0.37864	-0.46393	-0.66946	0.00448	-0.54332	0.18401	-0.26944	0.24139	0.30709	0.38857	0.23143	-0.06350	0.25318
	0.1213	0.0525	0.0024	0.9859	0.0198	0.4648	0.2796	0.3346	0.2151	0.1110	0.3555	0.8023	0.3107
	18	18	18	18	18	18	18	18	18	18	18	18	18
SO2	0.75127	0.67523	0.73330	0.52519	0.78480	0.37019	0.57116	0.50172	0.44426	0.49609	0.55138	-0.08942	0.29820
	0.0003	0.0021	0.0005	0.0252	0.0001	0.1305	0.0133	0.0339	0.0647	0.0363	0.0177	0.7242	0.2294
	18	18	18	18	18	18	18	18	18	18	18	18	18

	SF	NO3	CO	NO	NO2	O3	SO2
TA98P	0.71313	0.70324	0.66396	0.83251	0.59259	-0.37864	0.75127
	0.0009	0.0016	0.0027	0.0001	0.0096	0.1213	0.0003
	18	17	18	18	18	18	18
TA98M	0.83969	0.68520	0.50083	0.77972	0.39963	-0.46393	0.67523
	0.0001	0.0024	0.0343	0.0001	0.1004	0.0525	0.0021
	18	17	18	18	18	18	18
BAP	0.71558	0.57713	0.22512	0.67176	0.16270	-0.66946	0.73330
	0.0008	0.0153	0.3691	0.0023	0.5189	0.0024	0.0005
	18	17	18	18	18	18	18
COR	0.22603	0.37124	0.86192	0.75225	0.67239	0.00448	0.52519
	0.3671	0.1423	0.0001	0.0003	0.0022	0.9859	0.0252
	18	17	18	18	18	18	18
BO	0.72637	0.69133	0.43936	0.77128	0.35817	-0.54332	0.78480
	0.0006	0.0021	0.0681	0.0002	0.1444	0.0198	0.0001
	18	17	18	18	18	18	18

CORRELATION COEFFICIENTS / PROB > |R| UNDER H0:RHO=0 / NUMBER OF OBSERVATIONS

	SF	NO3	CO	NO	NO2	O3	SO2
PBF	0.09862	0.40748	0.76253	0.57718	0.89205	0.18401	0.37019
	0.6970	0.1045	0.0002	0.0121	0.0001	0.4648	0.1305
	18	17	18	18	18	18	18
BRF	0.44639	0.55993	0.78881	0.87660	0.78246	-0.26944	0.57116
	0.0633	0.0194	0.0001	0.0001	0.0001	0.2796	0.0133
	18	17	18	18	18	18	18
KF	0.10128	0.54401	0.72386	0.45750	0.92474	0.24139	0.50172
	0.6892	0.0240	0.0007	0.0563	0.0001	0.3346	0.0339
	18	17	18	18	18	18	18
ZNF	-0.05963	0.50399	0.60760	0.37468	0.74285	0.30709	0.44426
	0.8142	0.0391	0.0075	0.1255	0.0004	0.2151	0.0647
	18	17	18	18	18	18	18
FEF	-0.05255	0.50803	0.67066	0.31146	0.87231	0.38857	0.49609
	0.8359	0.0373	0.0023	0.2083	0.0001	0.1110	0.0363
	18	17	18	18	18	18	18
SIF	0.12180	0.47233	0.69959	0.37828	0.80553	0.23143	0.55138
	0.6302	0.0556	0.0012	0.1217	0.0001	0.3555	0.0177
	18	17	18	18	18	18	18
CLF	-0.30267	-0.14438	0.00752	-0.03706	0.05950	-0.06350	-0.08942
	0.2222	0.5804	0.9764	0.8839	0.8146	0.8023	0.7242
	18	17	18	18	18	18	18
NIF	0.22105	0.61873	0.11923	0.02979	0.64767	0.25318	0.29820
	0.3781	0.0081	0.6375	0.9066	0.0037	0.3107	0.2294
	18	17	18	18	18	18	18
SF	1.00000	0.64455	0.15196	0.48753	0.14107	-0.44444	0.57071
	0.0000	0.0052	0.5472	0.0401	0.5766	0.0646	0.0134
	18	17	18	18	18	18	18
NO3	0.64455	1.00000	0.31686	0.43096	0.59138	-0.15354	0.78859
	0.0052	0.0000	0.2153	0.0842	0.0124	0.5563	0.0002
	17	17	17	17	17	17	17
CO	0.15196	0.31686	1.00000	0.74134	0.70880	-0.01013	0.51216
	0.5472	0.2153	0.0000	0.0004	0.0010	0.9682	0.0298
	18	17	18	18	18	18	18
NO	0.48753	0.43096	0.74134	1.00000	0.55764	-0.53711	0.61918
	0.0401	0.0842	0.0004	0.0000	0.0162	0.0215	0.0061
	18	17	18	18	18	18	18
NO2	0.14107	0.59138	0.70880	0.55764	1.00000	0.05266	0.59471
	0.5766	0.0124	0.0010	0.0162	0.0000	0.8356	0.0092
	18	17	18	18	18	18	18

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	SF	NO3	CO	NO	NO2	O3	SO2
O3	-0.44444	-0.15354	-0.01013	-0.53711	0.05266	1.00000	-0.32031
	0.0646	0.5563	0.9682	0.0215	0.8356	0.0000	0.1950
	18	17	18	18	18	18	18
SO2	0.57071	0.78859	0.51216	0.61918	0.59471	-0.32031	1.00000
	0.0134	0.0002	0.0298	0.0061	0.0092	0.1950	0.0000
	18	17	18	18	18	18	18

VARIABLE	N	MEAN	STD DEV	SUM	MINIMUM	MAXIMUM
TA98P	18	27.34444442	13.76308883	492.19999955	7.70000000	58.39999998
TA98M	18	12.22222221	6.69245012	219.99999975	3.40000000	26.79999995
BAP	18	0.32222222	0.42086124	5.79999999	0.10000000	1.60000000
COR	18	0.87222222	0.36267885	15.69999999	0.30000000	1.60000000
BO	18	1.16111111	1.30435420	20.89999999	0.10000000	5.10000000
PBF	18	287.00000000	149.02348808	5166.00000000	86.00000000	667.00000000
BRF	18	52.00000000	34.38878066	936.00000000	6.00000000	125.00000000
KF	18	130.83333333	99.97249622	2355.00000000	36.00000000	406.00000000
ZNF	18	20.22222222	12.58643320	364.00000000	3.00000000	44.00000000
FEF	18	93.22222222	45.25035659	1678.00000000	20.00000000	180.00000000
SIF	18	205.50000000	99.44152878	3699.00000000	81.00000000	446.00000000
CLF	18	126.50000000	218.21151369	2277.00000000	18.00000000	971.00000000
NIF	18	2.66666667	1.37198868	48.00000000	2.00000000	7.00000000
SF	18	1350.55555556	959.90466575	24310.00000000	442.00000000	4540.00000000
NO3	18	7.04444444	3.38391041	126.79999998	3.50000000	16.50000000
CO	18	1.19999973	0.39704784	21.59999514	0.19999999	1.89999962
NO	18	2.27222183	1.60029593	40.89999300	0.09999996	6.29999924
NO2	18	3.17777745	1.02816555	57.19999409	1.89999962	5.00000000
O3	18	1.62222203	1.55433184	29.19999653	0	4.59999943
SO2	18	0.03888888	0.08498365	0.69999987	0	0.29999995

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	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
TA98P	1.00000 0.0000	0.95880 0.0001	0.33769 0.1705	0.30796 0.2138	0.45622 0.0570	-0.14393 0.5688	0.08990 0.7228	0.05364 0.8326	-0.13524 0.5926	0.00460 0.9855	-0.06952 0.7840	0.02155 0.9324	-0.01755 0.9449
TA98M	0.95880 0.0001	1.00000 0.0000	0.41939 0.0832	0.16604 0.5102	0.49250 0.0379	-0.26445 0.2889	-0.04611 0.8558	-0.08562 0.7355	-0.17367 0.4907	-0.11225 0.6574	-0.16946 0.5014	0.01194 0.9625	-0.13945 0.5810
BAP	0.33769 0.1705	0.41939 0.0832	1.00000 0.0000	0.18156 0.4709	0.95321 0.0001	-0.26383 0.2901	0.15729 0.5331	0.13137 0.6033	-0.03319 0.8960	-0.35950 0.1429	-0.12397 0.6241	0.54944 0.0182	0.18677 0.4580
COR	0.30796 0.2138	0.16604 0.5102	0.18156 0.4709	1.00000 0.0000	0.35446 0.1490	0.70265 0.0011	0.79283 0.0001	0.73934 0.0005	-0.03594 0.8874	0.60364 0.0080	0.61514 0.0066	-0.06136 0.8089	0.77235 0.0002
BO	0.45622 0.0570	0.49250 0.0379	0.95321 0.0001	0.35446 0.1490	1.00000 0.0000	-0.14965 0.5534	0.28353 0.2542	0.20042 0.4252	0.04606 0.8560	-0.30491 0.2186	-0.11295 0.6554	0.51501 0.0287	0.22571 0.3678
PBF	-0.14393 0.5688	-0.26445 0.2889	-0.26383 0.2901	0.70265 0.0011	-0.14965 0.5534	1.00000 0.0000	0.84410 0.0001	0.73014 0.0006	0.17409 0.4896	0.84866 0.0001	0.75510 0.0003	-0.11898 0.6382	0.63813 0.0044
BRF	0.08990 0.7228	-0.04611 0.8558	0.15729 0.5331	0.79283 0.0001	0.28353 0.2542	0.84410 0.0001	1.00000 0.0000	0.85075 0.0001	0.20290 0.4194	0.65601 0.0031	0.71360 0.0009	0.30259 0.2223	0.70691 0.0010
KF	0.05364 0.8326	-0.08562 0.7355	0.13137 0.6033	0.73934 0.0005	0.20042 0.4252	0.73014 0.0006	0.85075 0.0001	1.00000 0.0000	0.10793 0.6699	0.70146 0.0012	0.83221 0.0001	0.22757 0.3638	0.91863 0.0001
ZNF	-0.13524 0.5926	-0.17367 0.4907	-0.03319 0.8960	-0.03594 0.8874	0.04606 0.8560	0.17409 0.4896	0.20290 0.4194	0.10793 0.6699	1.00000 0.0000	0.01902 0.9403	0.06669 0.7926	0.44606 0.0635	-0.07721 0.7607
FEF	0.00460 0.9855	-0.11225 0.6574	-0.35950 0.1429	0.60364 0.0080	-0.30491 0.2186	0.84866 0.0001	0.65601 0.0031	0.70146 0.0012	0.01902 0.9403	1.00000 0.0000	0.90712 0.0001	-0.23023 0.3580	0.60387 0.0080
SIF	-0.06952 0.7840	-0.16946 0.5014	-0.12397 0.6241	0.61514 0.0066	-0.11295 0.6554	0.75510 0.0003	0.71360 0.0009	0.83221 0.0001	0.06669 0.7926	0.90712 0.0001	1.00000 0.0000	0.03781 0.8816	0.74978 0.0003
CLF	0.02155 0.9324	0.01194 0.9625	0.54944 0.0182	-0.06136 0.8089	0.51501 0.0287	-0.11898 0.6382	0.30259 0.2223	0.22757 0.3638	0.44606 0.0635	-0.23023 0.3580	0.03781 0.8816	1.00000 0.0000	0.01395 0.9562
NIF	-0.01755 0.9449	-0.13945 0.5810	0.18677 0.4580	0.77235 0.0002	0.22571 0.3678	0.63813 0.0044	0.70691 0.0010	0.91863 0.0001	-0.07721 0.7607	0.60387 0.0080	0.74978 0.0003	0.01395 0.9562	1.00000 0.0000
SF	-0.00948 0.9702	-0.06773 0.7895	0.41263 0.0888	0.01884 0.9409	0.45817 0.0559	0.04542 0.8580	0.42099 0.0819	0.31931 0.1965	0.61671 0.0064	-0.15333 0.5436	0.05262 0.8357	0.91598 0.0001	0.06616 0.7942
NO3	0.57359 0.0128	0.54898 0.0183	0.70681 0.0010	0.37109 0.1295	0.82110 0.0001	-0.12277 0.6274	0.23617 0.3454	0.19639 0.4348	0.06757 0.7899	-0.22576 0.3677	-0.08816 0.7280	0.26757 0.2831	0.22764 0.3636
CO	0.55631 0.0165	0.52842 0.0242	0.04224 0.8678	0.49836 0.0353	0.14993 0.5526	0.38285 0.1169	0.35844 0.1441	0.20851 0.4064	-0.20481 0.4149	0.30973 0.2110	0.18727 0.4568	-0.27334 0.2724	0.23756 0.3425
NO	0.32544 0.1876	0.25876 0.2998	0.21495 0.3917	0.75467 0.0003	0.35989 0.1424	0.62866 0.0052	0.69713 0.0013	0.50726 0.0317	-0.07765 0.7594	0.38188 0.1179	0.33233 0.1779	-0.02472 0.9224	0.51261 0.0296

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	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
NO2	0.04264	0.04085	-0.26388	0.49516	-0.24236	0.59937	0.36052	0.42562	-0.05869	0.64556	0.51792	-0.37459	0.45314
	0.8666	0.8721	0.2900	0.0367	0.3325	0.0086	0.1416	0.0782	0.8171	0.0038	0.0277	0.1256	0.0590
O3	-0.08513	-0.10636	-0.40365	-0.10841	-0.39356	0.16868	-0.04380	0.03076	0.36536	0.29733	0.28593	-0.24712	0.00644
	0.7370	0.6744	0.0967	0.6685	0.1061	0.5034	0.8630	0.9036	0.1360	0.2308	0.2501	0.3228	0.9798
SO2	0.37814	0.40175	0.40203	-0.07740	0.31162	-0.11333	0.17914	0.13582	0.16193	0.00068	0.12285	0.68627	-0.08408
	0.1218	0.0984	0.0982	0.7602	0.2081	0.6543	0.4769	0.5910	0.5209	0.9979	0.6272	0.0017	0.7401

	SF	NO3	CO	NO	NO2	O3	SO2
TA98P	-0.00948	0.57359	0.55631	0.32544	0.04264	-0.08513	0.37814
	0.9702	0.0128	0.0165	0.1876	0.8666	0.7370	0.1218
TA98M	-0.06773	0.54898	0.52842	0.25876	0.04085	-0.10636	0.40175
	0.7895	0.0183	0.0242	0.2998	0.8721	0.6744	0.0984
BAP	0.41263	0.70681	0.04224	0.21495	-0.26388	-0.40365	0.40203
	0.0888	0.0010	0.8678	0.3917	0.2900	0.0967	0.0982
COR	0.01884	0.37109	0.49836	0.75467	0.49516	-0.10841	-0.07740
	0.9409	0.1295	0.0353	0.0003	0.0367	0.6685	0.7602
BO	0.45817	0.82110	0.14993	0.35989	-0.24236	-0.39356	0.31162
	0.0559	0.0001	0.5526	0.1424	0.3325	0.1061	0.2081
PBF	0.04542	-0.12277	0.38285	0.62866	0.59937	0.16868	-0.11333
	0.8580	0.6274	0.1169	0.0052	0.0086	0.5034	0.6543
BRF	0.42099	0.23617	0.35844	0.69713	0.36052	-0.04380	0.17914
	0.0819	0.3454	0.1441	0.0013	0.1416	0.8630	0.4769
KF	0.31931	0.19639	0.20851	0.50726	0.42562	0.03076	0.13582
	0.1965	0.4348	0.4064	0.0317	0.0782	0.9036	0.5910
ZNF	0.61671	0.06757	-0.20481	-0.07765	-0.05869	0.36536	0.16193
	0.0064	0.7899	0.4149	0.7594	0.8171	0.1360	0.5209
FEF	-0.15333	-0.22576	0.30973	0.38188	0.64556	0.29733	0.00068
	0.5436	0.3677	0.2110	0.1179	0.0038	0.2308	0.9979
SIF	0.05262	-0.08816	0.18727	0.33233	0.51792	0.28593	0.12285
	0.8357	0.7280	0.4568	0.1779	0.0277	0.2501	0.6272
CLF	0.91598	0.26757	-0.27334	-0.02472	-0.37459	-0.24712	0.68627
	0.0001	0.2831	0.2724	0.9224	0.1256	0.3228	0.0017
NIF	0.06616	0.22764	0.23756	0.51261	0.45314	0.00644	-0.08408
	0.7942	0.3636	0.3425	0.0296	0.0590	0.9798	0.7401

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / N = 18

	SF	NO3	CO	NO	NO2	O3	SO2
SF	1.00000 0.0000	0.33200 0.1783	-0.29567 0.2336	-0.00496 0.9844	-0.38569 0.1139	-0.14233 0.5732	0.49893 0.0351
NO3	0.33200 0.1783	1.00000 0.0000	0.27495 0.2695	0.25388 0.3094	-0.34866 0.1562	0.01501 0.9529	0.12659 0.6167
CO	-0.29567 0.2336	0.27495 0.2695	1.00000 0.0000	0.71100 0.0009	0.46975 0.0492	0.10103 0.6900	0.00000 1.0000
NO	-0.00496 0.9844	0.25388 0.3094	0.71100 0.0009	1.00000 0.0000	0.44577 0.0637	-0.15038 0.5514	0.01706 0.9464
NO2	-0.38569 0.1139	-0.34866 0.1562	0.46975 0.0492	0.44577 0.0637	1.00000 0.0000	0.00732 0.9770	-0.11744 0.6426
O3	-0.14233 0.5732	0.01501 0.9529	0.10103 0.6900	-0.15038 0.5514	0.00732 0.9770	1.00000 0.0000	-0.16279 0.5187
SO2	0.49893 0.0351	0.12659 0.6167	0.00000 1.0000	0.01706 0.9464	-0.11744 0.6426	-0.16279 0.5187	1.00000 0.0000

VARIABLE	N	MEAN	STD DEV	SUM	MINIMUM	MAXIMUM
TA98P	18	14.07777777	8.17554139	253.39999978	0.60000000	28.39999998
TA98M	18	7.07222222	4.26391225	127.29999995	0.40000000	14.20000000
BAP	18	0.13888889	0.06978023	2.50000000	0.10000000	0.30000000
COR	18	0.32222222	0.25565500	5.80000000	0.10000000	0.90000000
BO	18	0.48333333	0.31482955	8.70000000	0.10000000	1.20000000
PBF	18	228.26611111	202.03637556	4108.79000000	17.00000000	771.00000000
BRF	18	44.46111111	35.33125826	800.30000000	7.00000000	130.00000000
KF	18	139.52777778	115.77552218	2511.50000000	37.00000000	437.00000000
ZNF	18	24.73888889	15.52118624	445.30000000	4.00000000	74.00000000
FEF	18	174.22777778	125.44174090	3136.10000000	17.00000000	421.00000000
SIF	18	450.95555556	334.41987385	8117.20000000	79.00000000	1220.00000000
CLF	18	264.21111111	217.83477519	4755.80000000	19.00000000	893.00000000
NIF	18	16.85555555	12.60598468	303.39999998	2.00000000	48.00000000
GF	18	1968.42222222	1153.93953280	35431.60000000	541.00000000	5790.00000000
NO3	18	7.81111111	5.65174506	140.59999998	3.00000000	24.50000000
NO	9	2.71111094	1.72224005	24.39999843	0.29999995	6.39999962
NO2	15	2.69999955	1.49092499	40.49999332	1.19999981	6.19999981
O3	15	1.77333314	1.20621393	26.59999704	0	3.19999981
SO2	15	1.35333300	1.14633970	20.29999506	0.29999995	4.29999924

CORRELATIONS OF MARTINEZ EPISODE DATA, pp. 29-33

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
TA98P	1.00000 0.0000 18	0.92935 0.0001 18	0.28825 0.2461 18	-0.01129 0.9645 18	0.48961 0.0392 18	0.13388 0.5964 18	0.33461 0.1747 18	0.18025 0.4742 18	0.13720 0.5872 18	-0.02539 0.9204 18	-0.16409 0.5153 18	-0.32350 0.1904 18	0.10007 0.6928 18
TA98M	0.92935 0.0001 18	1.00000 0.0000 18	0.32610 0.1866 18	0.15817 0.5308 18	0.46062 0.0544 18	0.33824 0.1698 18	0.47603 0.0458 18	0.34902 0.1557 18	0.33729 0.1711 18	0.16881 0.5031 18	0.01959 0.9385 18	-0.26315 0.2914 18	0.12028 0.6345 18
BAP	0.28825 0.2461 18	0.32610 0.1866 18	1.00000 0.0000 18	0.70710 0.0010 18	0.83451 0.0001 18	0.57460 0.0126 18	0.72955 0.0006 18	0.55650 0.0165 18	0.34666 0.1587 18	0.43439 0.0717 18	0.36029 0.1419 18	-0.25018 0.3167 18	-0.23398 0.3501 18
COR	-0.01129 0.9645 18	0.15817 0.5308 18	0.70710 0.0010 18	1.00000 0.0000 18	0.63339 0.0048 18	0.84473 0.0001 18	0.84957 0.0001 18	0.79294 0.0001 18	0.54174 0.0202 18	0.76834 0.0002 18	0.72613 0.0006 18	-0.01103 0.9653 18	-0.19023 0.4496 18
BO	0.48961 0.0392 18	0.46062 0.0544 18	0.83451 0.0001 18	0.63339 0.0048 18	1.00000 0.0000 18	0.45616 0.0571 18	0.67388 0.0022 18	0.46935 0.0494 18	0.23343 0.3512 18	0.27588 0.2678 18	0.21759 0.3858 18	-0.23724 0.3432 18	-0.33828 0.1697 18
PBF	0.13388 0.5964 18	0.33824 0.1698 18	0.57460 0.0126 18	0.84473 0.0001 18	0.45616 0.0571 18	1.00000 0.0000 18	0.92944 0.0001 18	0.92958 0.0001 18	0.74960 0.0003 18	0.90022 0.0001 18	0.74867 0.0004 18	-0.05122 0.8400 18	0.18641 0.4589 18
BRF	0.33461 0.1747 18	0.47603 0.0458 18	0.72955 0.0006 18	0.84957 0.0001 18	0.67388 0.0022 18	0.92944 0.0001 18	1.00000 0.0000 18	0.84864 0.0001 18	0.68574 0.0017 18	0.76972 0.0002 18	0.59950 0.0085 18	-0.12210 0.6294 18	0.06953 0.7840 18
KF	0.18025 0.4742 18	0.34902 0.1557 18	0.55650 0.0165 18	0.79294 0.0001 18	0.46935 0.0494 18	0.92958 0.0001 18	0.84864 0.0001 18	1.00000 0.0000 18	0.65671 0.0031 18	0.80442 0.0001 18	0.69831 0.0013 18	0.00024 0.9993 18	0.09793 0.6991 18
ZNF	0.13720 0.5872 18	0.33729 0.1711 18	0.34666 0.1587 18	0.54174 0.0202 18	0.23343 0.3512 18	0.74960 0.0003 18	0.68574 0.0017 18	0.65671 0.0031 18	1.00000 0.0000 18	0.72305 0.0007 18	0.54970 0.0181 18	-0.18941 0.4516 18	0.49618 0.0362 18
FEF	-0.02539 0.9204 18	0.16881 0.5031 18	0.43439 0.0717 18	0.76834 0.0002 18	0.27588 0.2678 18	0.90022 0.0001 18	0.76972 0.0002 18	0.80442 0.0001 18	0.72305 0.0007 18	1.00000 0.0000 18	0.90809 0.0001 18	-0.11814 0.6406 18	0.33937 0.1683 18
SIF	-0.16409 0.5153 18	0.01959 0.9385 18	0.36029 0.1419 18	0.72613 0.0006 18	0.21759 0.3858 18	0.74867 0.0004 18	0.59950 0.0085 18	0.69831 0.0013 18	0.54970 0.0181 18	0.90809 0.0001 18	1.00000 0.0000 18	0.15372 0.5425 18	0.14030 0.5787 18
CLF	-0.32350 0.1904 18	-0.26315 0.2914 18	-0.25018 0.3167 18	-0.01103 0.9653 18	-0.23724 0.3432 18	-0.05122 0.8400 18	-0.12210 0.6294 18	0.00024 0.9993 18	-0.18941 0.4516 18	-0.11814 0.6406 18	0.15372 0.5425 18	1.00000 0.0000 18	-0.27728 0.2653 18
NIF	0.10007 0.6928 18	0.12028 0.6345 18	-0.23398 0.3501 18	-0.19023 0.4496 18	-0.33828 0.1697 18	0.18641 0.4589 18	0.06953 0.7840 18	0.09793 0.6991 18	0.49618 0.0362 18	0.33937 0.1683 18	0.14030 0.5787 18	-0.27728 0.2653 18	1.00000 0.0000 18

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	TA98P	TA98M	BAP	COR	BO	PBF	BRF	KF	ZNF	FEF	SIF	CLF	NIF
SF	0.51471	0.51491	0.04099	-0.02761	0.18682	0.15363	0.23184	0.33801	0.36622	0.01413	-0.01806	0.08135	0.23057
	0.0288	0.0288	0.8717	0.9134	0.4579	0.5428	0.3546	0.1701	0.1350	0.9556	0.9433	0.7483	0.3573
	18	18	18	18	18	18	18	18	18	18	18	18	18
NO3	0.63242	0.56885	0.15694	-0.00873	0.46691	-0.10536	0.10681	0.06973	0.06362	-0.26846	-0.23405	0.06398	-0.23346
	0.0049	0.0138	0.5340	0.9726	0.0508	0.6773	0.6731	0.7834	0.8020	0.2814	0.3499	0.8009	0.3512
	18	18	18	18	18	18	18	18	18	18	18	18	18
CO													
	0	0	0	0	0	0	0	0	0	0	0	0	0
NO	0.55212	0.48690	0.82799	0.51547	0.73964	0.36537	0.55405	0.17045	0.43112	0.13157	-0.01367	-0.31045	-0.14897
	0.1232	0.1838	0.0058	0.1555	0.0227	0.3336	0.1217	0.6611	0.2466	0.7358	0.9722	0.4162	0.7021
	9	9	9	9	9	9	9	9	9	9	9	9	9
NO2	0.29052	0.51279	0.41615	0.74490	0.33341	0.93532	0.83513	0.85814	0.74557	0.93257	0.89792	-0.16144	0.32692
	0.2935	0.0506	0.1229	0.0014	0.2246	0.0001	0.0001	0.0001	0.0014	0.0001	0.0001	0.5654	0.2343
	15	15	15	15	15	15	15	15	15	15	15	15	15
O3	-0.47051	-0.53725	-0.48545	-0.37138	-0.62129	-0.24779	-0.45195	-0.24388	-0.31080	-0.01512	-0.04239	-0.15703	0.28110
	0.0767	0.0389	0.0666	0.1729	0.0134	0.3732	0.0908	0.3811	0.2595	0.9573	0.8808	0.5762	0.3102
	15	15	15	15	15	15	15	15	15	15	15	15	15
SO2	0.73863	0.58663	0.41270	-0.01688	0.52023	-0.14223	0.14874	-0.06508	-0.04904	-0.32131	-0.31231	0.15680	-0.22241
	0.0017	0.0215	0.1263	0.9524	0.0468	0.6131	0.5968	0.8177	0.8622	0.2429	0.2571	0.5768	0.4256
	15	15	15	15	15	15	15	15	15	15	15	15	15

	SF	NO3	CO	NO	NO2	O3	SO2
TA98P	0.51471	0.63242		0.55212	0.29052	-0.47051	0.73863
	0.0288	0.0049		0.1232	0.2935	0.0767	0.0017
	18	18	0	9	15	15	15
TA98M	0.51491	0.56885		0.48690	0.51279	-0.53725	0.58663
	0.0288	0.0138		0.1838	0.0506	0.0389	0.0215
	18	18	0	9	15	15	15
BAP	0.04099	0.15694		0.82799	0.41615	-0.48545	0.41270
	0.8717	0.5340		0.0058	0.1229	0.0666	0.1263
	18	18	0	9	15	15	15
COR	-0.02761	-0.00873		0.51547	0.74490	-0.37138	-0.01688
	0.9134	0.9726		0.1555	0.0014	0.1729	0.9524
	18	18	0	9	15	15	15
BO	0.18682	0.46691		0.73964	0.33341	-0.62129	0.52023
	0.4579	0.0508		0.0227	0.2246	0.0134	0.0468
	18	18	0	9	15	15	15

STATION=7996

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	SF	NO3	CO	NO	NO2	O3	SO2
PBF	0.15363	-0.10536		0.36537	0.93532	-0.24779	-0.14223
	0.5428	0.6773		0.3336	0.0001	0.3732	0.6131
	18	18	0	9	15	15	15
BRF	0.23184	0.10681		0.55405	0.83513	-0.45195	0.14874
	0.3546	0.6731		0.1217	0.0001	0.0908	0.5968
	18	18	0	9	15	15	15
KF	0.33801	0.06973		0.17045	0.85814	-0.24388	-0.06508
	0.1701	0.7834		0.6611	0.0001	0.3811	0.8177
	18	18	0	9	15	15	15
ZNF	0.36622	0.06362		0.43112	0.74557	-0.31080	-0.04904
	0.1350	0.8020		0.2466	0.0014	0.2595	0.8622
	18	18	0	9	15	15	15
FEF	0.01413	-0.26846		0.13157	0.93257	-0.01512	-0.32131
	0.9556	0.2814		0.7358	0.0001	0.9573	0.2429
	18	18	0	9	15	15	15
SIF	-0.01806	-0.23405		-0.01367	0.89792	-0.04239	-0.31231
	0.9433	0.3499		0.9722	0.0001	0.8808	0.2571
	18	18	0	9	15	15	15
CLF	0.08135	0.06398		-0.31045	-0.16144	-0.15703	0.15680
	0.7483	0.8009		0.4162	0.5654	0.5762	0.5768
	18	18	0	9	15	15	15
NIF	0.23057	-0.23346		-0.14897	0.32692	0.28110	-0.22241
	0.3573	0.3512		0.7021	0.2343	0.3102	0.4256
	18	18	0	9	15	15	15
SF	1.00000	0.66667		-0.05736	0.17244	-0.41658	0.52203
	0.0000	0.0025		0.8835	0.5389	0.1224	0.0459
	18	18	0	9	15	15	15
NO3	0.66667	1.00000		0.22436	-0.09255	-0.58411	0.79808
	0.0025	0.0000		0.5617	0.7429	0.0222	0.0004
	18	18	0	9	15	15	15
CO							
	0	0	0	0	0	0	0
NO	-0.05736	0.22436		1.00000	0.19817	-0.30163	0.58598
	0.8835	0.5617		0.0000	0.6093	0.4302	0.0973
	9	9	0	9	9	9	9
NO2	0.17244	-0.09255		0.19817	1.00000	-0.09374	-0.16884
	0.5389	0.7429		0.6093	0.0000	0.7397	0.5475
	15	15	0	9	15	15	15

CORRELATION COEFFICIENTS / PROB > |R| UNDER HO:RHO=0 / NUMBER OF OBSERVATIONS

	SF	NO3	CO	NO	NO2	O3	SO2
O3	-0.41658	-0.58411		-0.30163	-0.09374	1.00000	-0.56403
	0.1224	0.0222		0.4302	0.7397	0.0000	0.0285
	15	15	0	9	15	15	15
SO2	0.52203	0.79808		0.58598	-0.16884	-0.56403	1.00000
	0.0459	0.0004		0.0973	0.5475	0.0285	0.0000
	15	15	0	9	15	15	15

APPENDIX IV: COMPLETE DATA SET FOR CONTRA COSTA SEASONAL COMPOSITES NOVEMBER 1979 - OCTOBER 1984

STATION 7430 = PITTSBURG, STATION 7433 = RICHMOND, STATION 7440 = CONCORD, PERIOD 801 = NOVEMBER 1979 - FEBRUARY 1980, PERIOD 802 = MARCH 1980 - JUNE 1980, ETC.

STATION=7430

OBS	STATION	PERIOD	MASS	LEAD	SO4	NO3	ORGANICS	BKF	BAP	BGP	CDR	BO	TA98P	TA98M	TA98NRP	TA98NRM
1	7430	801	75	0.43	8.0	11.4	4.4	0.3	0.4	2.1	1.2	0.6	5.4	3.4	8.1	4.8
2	7430	802	62	0.17	5.9	3.9	1.5	0.1	0.1	0.4	0.2	0.1	1.1	1.0	2.1	1.0
3	7430	803	91	0.29	9.5	6.6	2.6	0.1	0.1	0.4	0.2	0.1	4.4	3.7	1.8	1.3
4	7430	811	85	0.39	9.4	12.2	5.5	0.4	0.5	2.4	1.2	1.0	8.8	4.1	4.5	1.7
5	7430	812	59	0.16	6.2	4.0	3.1	0.1	0.1	0.4	0.3	0.1	1.8	1.8	1.0	0.5
6	7430	813	75	0.19	8.0	6.3	4.4	0.1	0.1	0.3	0.2	0.1	5.1	3.2	2.2	0.9
7	7430	821	58	0.35	5.7	9.4	5.9	0.3	0.5	1.8	0.9	1.3	13.2	9.7	8.0	3.2
8	7430	822	62	0.18	5.7	4.6	2.6	0.1	0.1	0.3	0.2	0.1	4.5	4.1	3.6	1.6
9	7430	823	58	0.20	6.0	4.9	3.5	0.1	0.2	1.0	1.0	0.2	12.7	5.4	7.6	2.9
10	7430	831	50	0.25	4.9	8.8	5.0	0.5	0.6	3.3	2.2	2.4	11.3	6.0	7.3	3.7
11	7430	832	44	0.11	5.0	2.8	0.9	0.0	0.0	0.4	0.4	0.0	8.1	3.6	2.3	0.8
12	7430	833	58	0.17	6.5	5.2	1.3	0.0	0.1	0.5	0.2	0.2	14.8	7.4	6.2	1.7
13	7430	841	56	0.24	6.1	7.4	4.6	0.3	0.6	2.3	1.0	1.9	30.1	14.8	8.9	3.2
14	7430	842	57	0.15	6.0	4.2	2.9	0.0	0.1	0.3	0.2	0.2	16.1	8.9	4.8	1.5
15	7430	843	65	0.14	.	.	2.3	0.1	0.1	0.6	0.5	0.1	12.4	8.1	3.9	1.7

STATION=7433

OBS	STATION	PERIOD	MASS	LEAD	SO4	NO3	ORGANICS	BKF	BAP	BGP	CDR	BO	TA98P	TA98M	TA98NRP	TA98NRM
16	7433	801	64	0.57	9.1	9.2	5.6	0.3	0.3	2.7	1.5	0.5	12.3	6.8	6.9	3.3
17	7433	802	52	0.28	6.7	2.3	2.3	0.1	0.1	0.5	0.4	0.1	3.4	3.0	1.6	0.9
18	7433	803	60	0.30	8.5	4.3	3.1	0.1	0.1	0.4	0.3	0.1	4.0	2.3	2.3	1.0
19	7433	811	76	0.43	12.2	10.1	5.4	0.3	0.3	2.5	1.4	0.7	12.7	6.8	3.6	7.0
20	7433	812	65	0.21	6.9	3.5	3.6	0.1	0.1	0.6	0.4	0.1	2.4	2.8	1.8	0.6
21	7433	813	55	0.20	6.1	3.6	4.0	0.1	0.1	0.5	0.3	0.1	19.1	4.7	7.5	3.6
22	7433	821	63	0.43	5.2	6.9	5.1	0.4	0.5	2.6	1.3	1.3	11.3	5.7	6.8	3.2
23	7433	822	53	0.22	6.5	3.8	3.4	0.1	0.1	0.4	0.3	0.1	6.0	4.9	3.3	2.6
24	7433	823	49	0.20	6.5	2.9	3.5	0.1	0.1	1.1	1.2	0.1	3.3	2.6	2.6	0.6
25	7433	831	51	0.32	7.0	5.2	5.8	0.6	0.9	4.8	3.5	1.7	14.7	6.6	7.8	3.4
26	7433	832	42	0.16	6.5	2.5	1.3	0.1	0.1	1.1	1.1	0.2	11.8	4.9	5.5	1.6
27	7433	833	55	0.18	7.6	3.8	1.9	0.1	0.1	0.7	0.3	0.3	14.7	6.0	6.4	1.9
28	7433	841	52	0.28	6.6	5.1	5.9	0.4	0.7	3.3	1.4	2.0	27.4	15.2	9.4	3.9
29	7433	842	53	0.14	7.3	3.3	3.1	0.1	0.1	0.6	0.4	0.2	17.5	10.0	7.1	2.3
30	7433	843	69	0.15	.	.	2.1	0.1	0.1	0.8	0.7	0.1	11.1	6.3	3.0	1.8

STATION=7440

OBS	STATION	PERIOD	MASS	LEAD	SO4	NO3	ORGANICS	BKF	BAP	BGP	CDR	BO	TA98P	TA98M	TA98NRP	TA98NRM
31	7440	801	73	0.69	7.7	11.2	10.7	0.6	1.3	4.1	1.9	3.3	4.8	2.7	7.7	2.4
32	7440	802	38	0.23	3.7	2.7	1.5	0.1	0.1	0.5	0.5	0.1	1.2	1.5	3.0	1.4
33	7440	803	60	0.44	6.9	7.0	3.5	0.1	0.1	0.7	0.4	0.2	7.4	5.0	3.4	1.7
34	7440	811	90	0.73	7.4	10.1	10.5	0.6	1.2	3.9	1.8	1.9	14.4	7.5	9.0	1.6
35	7440	812	45	0.24	4.4	4.5	3.4	0.1	0.1	0.9	0.5	0.1	2.4	2.5	1.6	0.9
36	7440	813	52	0.27	6.1	6.1	4.0	0.1	0.1	0.6	0.3	0.2	7.4	4.2	3.4	1.2
37	7440	821	61	0.50	4.4	8.3	10.7	0.6	1.5	4.0	1.8	2.9	5.5	3.5	3.0	1.7
38	7440	822	43	0.24	4.6	4.7	2.6	0.1	0.1	0.7	0.4	0.2	6.6	4.7	3.0	3.2
39	7440	823	39	0.24	4.9	4.4	2.9	0.1	0.1	1.1	1.1	0.1	17.1	5.4	4.6	1.7
40	7440	831	56	0.42	4.1	6.9	10.6	1.0	2.3	7.5	4.8	4.5	14.8	5.2	10.6	3.0
41	7440	832	32	0.17	4.4	2.8	1.0	0.1	0.1	1.1	1.2	0.1	11.0	6.0	5.0	1.4
42	7440	833	45	0.20	5.5	5.0	1.2	0.1	0.1	0.7	0.3	0.2	17.8	7.7	8.4	2.4

STATION=7440

OBS	STATION	PERIOD	MASS	LEAD	SO4	NO3	ORGANICS	BKF	BAP	BGP	COR	BD	TA98P	TA98N	TA98NRP	TA98NRM
43	7440	841	50	0.29	4.5	6.3	8.1	0.6	1.6	4.5	1.6	2.5	22.6	12.2	11.4	2.8
44	7440	842	39	0.23	4.8	4.6	2.8	0.0	0.1	0.5	0.3	0.2	19.2	12.0	4.6	1.6
45	7440	843	46	0.16	.	.	2.2	0.1	0.1	0.8	0.8	0.3	14.7	8.8	4.3	1.5

APPENDIX V:

LINEAR REGRESSION SLOPES OF COMPOSITE AEROSOL POLLUTANT DATA, 1979-1984. YEAR VERSUS SEASON AND ANNUAL AVERAGE

Variable	Season	Slope	P Value	Variable	Season	Slope	P Value
Pb	Winter	-0.08	≤0.001**	COR	Winter	0.2	0.65
	Spring	-0.01	0.20		Spring	0.04	0.71
	Summer	-0.04	0.03**		Summer	0.07	0.60
	Annual	-0.04	0.001**		Annual	0.09	0.50
NO_3	Winter	-1.3	0.01**	BZO	Winter	0.3	0.15
	Spring	0.08	0.77		Spring	0.02	0.16
	Summer	-0.5	0.19		Summer	0.02	0.28
	Annual	-0.5	0.05**		Annual	0.1	0.13
TSP	Winter	-7	0.10				
	Spring	-2	0.42				
	Summer	-3	0.34				
	Annual	-4	0.12				
SO_4	Winter	-0.9	0.16				
	Spring	0.05	0.63				
	Summer	-0.6	0.24				
	Annual	-0.5	0.11				
Organics	Winter	-0.1	0.36				
	Spring	0.0	0.99				
	Summer	-0.4	0.21				
	Annual	-0.2	0.42				
BAP	Winter	5	0.14				
	Spring	0.0	0.56				
	Summer	0.0	1.00				
	Annual	0.04	0.12				

** Slope different than zero at the P ≤0.05 level of significance.